4. Industrial Processes and Product Use

The Industrial Processes and Product Use (IPPU) chapter includes greenhouse gas emissions occurring from industrial processes and from the use of greenhouse gases in products. This chapter includes sources of emissions formerly represented in the 'Industrial Processes' and 'Solvent and Other Product Use' chapters in prior versions of this report. The industrial processes and product use categories included in this chapter are presented in Figure 4-1.

Greenhouse gas emissions are produced as the by-products of various non-energy-related industrial activities. That is, these emissions are produced either from an industrial process itself, and are not directly a result of energy consumed during the process. For example, raw materials can be chemically or physically transformed from one state to another. This transformation can result in the release of greenhouse gases such as carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O). The processes included in this chapter include iron and steel production and metallurgical coke production, cement production, lime production, other process uses of carbonates (e.g., flux stone, flue gas desulfurization, and glass manufacturing), ammonia production and urea consumption, petrochemical production, aluminum production, soda ash production and use, titanium dioxide production, CO_2 consumption, ferroalloy production, glass production, zinc production, phosphoric acid production, lead production, silicon carbide production and consumption, nitric acid production, and adipic acid production.

In addition, greenhouse gases are often used in products or by end-consumers. These gases include industrial sources of man-made compounds such as hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), sulfur hexafluoride (SF_6), nitrogen trifluoride (NF_3), and nitrous oxide (N_2O). The present contribution of HFCs, PFCs, SF_6, and NF_3 gases to the radiative forcing effect of all anthropogenic greenhouse gases is small; however, because of their extremely long lifetimes, many of them will continue to accumulate in the atmosphere as long as emissions continue. In addition, many of these gases have high global warming potentials; SF_6 is the most potent greenhouse gas the Intergovernmental Panel on Climate Change (IPCC) has evaluated. Use of HFCs is growing rapidly since they are the primary substitutes for ozone depleting substances (ODSs), which are being phased-out under the Montreal Protocol on Substances that Deplete the Ozone Layer. HFCs, PFCs, SF_6, and NF_3 are employed and emitted by a number of other industrial sources in the United States such as aluminum production, HCFC-22 production, semiconductor manufacture, electric power transmission and distribution, and magnesium metal production and processing. N_2O is emitted by the production of adipic acid and nitric acid, semiconductor manufacturing, end-consumers in product uses through the administration of anesthetics, and by industry as a propellant in aerosol products.

In 2013, IPPU generated emissions of 359.1 million metric tons of CO_2 equivalent (MMT CO_2 Eq.),[147] or 5.4 percent of total U.S. greenhouse gas emissions. Carbon dioxide emissions from all industrial processes were 163.0 MMT CO_2 Eq. (162,979 kt) in 2013, or 3.0 percent of total U.S. CO_2 emissions. Methane emissions from industrial processes resulted in emissions of approximately 0.8 MMT CO_2 Eq. (32 kt) in 2013, which was less than 1 percent of U.S. CH_4 emissions. N_2O emissions from IPPU were 19.1 MMT CO_2 Eq. (64 kt) in 2013, or 5.4 percent of total U.S. N_2O emissions. In 2013 combined emissions of HFCs, PFCs, SF_6, and NF_3 totaled 176.3 MMT CO_2 Eq. Total emissions from IPPU in 2013 were 5.0 percent more than 1990 emissions. Indirect greenhouse gas emissions also result from IPPU, and are presented in Table 4-106 in kilotons (kt).

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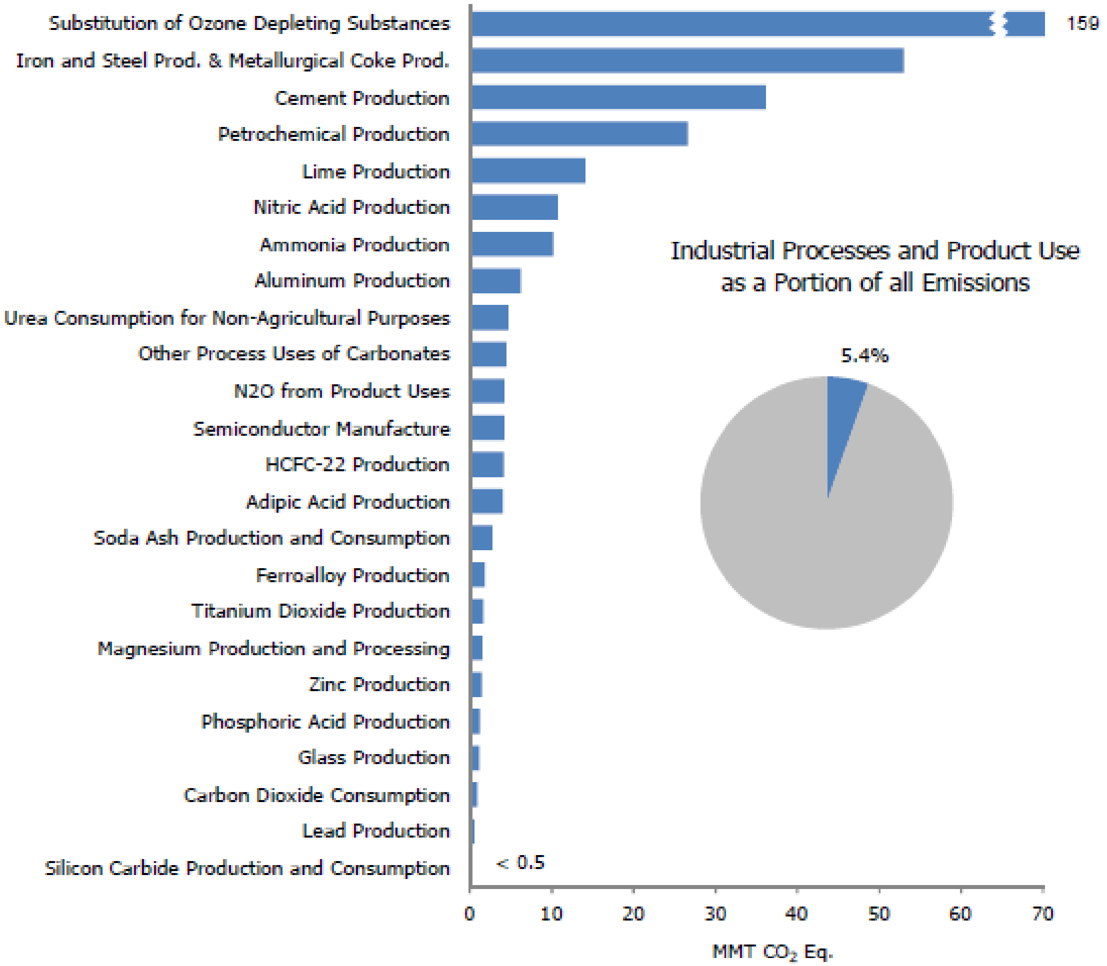
[147] Following the revised reporting requirements under the UNFCCC, this Inventory report presents CO_2 equivalent values based on the *IPCC Fourth Assessment Report* (AR4) GWP values. See the Introduction chapter for more information.

Figure 4-1: 2013 Industrial Processes and Product Use Chapter Greenhouse Gas Sources

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.



The increase in overall IPPU emissions since 1990 reflects a range of emission trends among the emission sources. Emissions resulting from most types of metal production have declined significantly since 1990, largely due to production shifting to other countries, but also due to transitions to less-emissive methods of production (in the case of iron and steel) and to improved practices (in the case of PFC emissions from aluminum production). Emissions from mineral sources have either increased or not changed significantly since 1990 but largely track economic cycles, while CO_2 and CH_4 emissions from chemical sources have either decreased or not changed significantly. HFC emissions from the substitution of ozone depleting substances have increased drastically since 1990, while the emission trends of HFCs, PFCs, SF_6, and NF_3 from other sources are mixed. N_2O emissions from the production of adipic and nitric acid have decreased, while N_2O emissions from product uses has remained nearly constant over time. Trends are explained further within each emission source category throughout the chapter.

Table 4-1 summarizes emissions for the IPPU chapter in MMT CO_2 Eq. using *IPCC Fourth Assessment Report* (AR4) GWP values, following the requirements of the revised UNFCCC reporting guidelines for national inventories (IPCC 2007).[148] Unweighted native gas emissions in kt are also provided in Table 4-2. The source descriptions that follow in the chapter are presented in the order as reported to the UNFCCC in the common

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[148] See < http://unfccc.int/resource/docs/2013/cop19/eng/10a03.pdf >.

reporting format tables, corresponding generally to: mineral products, chemical production, metal production, and emissions from the uses of HFCs, PFCs, SF_6, and NF_3.

Table 4-1: Emissions from Industrial Processes and Product Use (MMT CO_2 Eq.)

Gas/Source	1990	2005	2009	2010	2011	2012	2013
CO_2	**207.2**	**191.1**	**141.1**	**165.7**	**169.7**	**166.4**	**163.0**
Iron and Steel Production & Metallurgical Coke Production	99.8	66.7	43.0	55.7	60.0	54.3	52.3
Iron and Steel Production	*97.3*	*64.6*	*42.1*	*53.7*	*58.6*	*53.8*	*50.5*
Metallurgical Coke Production	*2.5*	*2.0*	*1.0*	*2.1*	*1.4*	*0.5*	*1.8*
Cement Production	33.3	45.9	29.4	31.3	32.0	35.1	36.1
Petrochemical Production	21.6	28.1	23.7	27.4	26.4	26.5	26.5
Lime Production	11.7	14.6	11.4	13.4	14.0	13.7	14.1
Ammonia Production	13.0	9.2	8.5	9.2	9.3	9.4	10.2
Urea Consumption for Non-Agricultural Purposes	3.8	3.7	3.4	4.7	4.0	4.4	4.7
Other Process Uses of Carbonates	4.9	6.3	7.6	9.6	9.3	8.0	4.4
Aluminum Production	6.8	4.1	3.0	2.7	3.3	3.4	3.3
Soda Ash Production and Consumption	2.7	2.9	2.5	2.6	2.6	2.7	2.7
Ferroalloy Production	2.2	1.4	1.5	1.7	1.7	1.9	1.8
Titanium Dioxide Production	1.2	1.8	1.6	1.8	1.7	1.5	1.6
Zinc Production	0.6	1.0	0.9	1.2	1.3	1.5	1.4
Phosphoric Acid Production	1.6	1.4	1.0	1.1	1.2	1.1	1.2
Glass Production	1.5	1.9	1.0	1.5	1.3	1.2	1.2
Carbon Dioxide Consumption	1.5	1.4	1.8	1.2	0.8	0.8	0.9
Lead Production	0.5	0.6	0.5	0.5	0.5	0.5	0.5
Silicon Carbide Production and Consumption	0.4	0.2	0.1	0.2	0.2	0.2	0.2
Magnesium Production and Processing	+	+	+	+	+	+	+
CH_4	**1.4**	**1.0**	**0.5**	**0.7**	**0.8**	**0.8**	**0.8**
Iron and Steel Production & Metallurgical Coke Production	1.1	0.9	0.4	0.6	0.7	0.7	0.7
Iron and Steel Production	*1.1*	*0.9*	*0.4*	*0.6*	*0.7*	*0.7*	*0.7*
Metallurgical Coke Production	+	+	+	+	+	+	+
Petrochemical Production	0.2	0.1	+	0.1	+	0.1	0.1
Ferroalloy Production	+	+	+	+	+	+	+
Silicon Carbide Production and Consumption	+	+	+	+	+	+	+
N_2O	**31.6**	**22.8**	**16.7**	**20.1**	**25.5**	**20.4**	**19.1**
Nitric Acid Production	12.1	11.3	9.6	11.5	10.9	10.5	10.7
N_2O from Product Uses	4.2	4.2	4.2	4.2	4.2	4.2	4.2
Adipic Acid Production	15.2	7.1	2.7	4.2	10.2	5.5	4.0
Semiconductor Manufacturing	+	0.1	0.1	0.1	0.2	0.2	0.2
HFCs	**46.6**	**131.4**	**142.9**	**152.6**	**157.4**	**159.2**	**163.0**
Substitution of Ozone Depleting Substances[a]	0.3	111.1	136.0	144.4	148.4	153.5	158.6
HCFC-22 Production	46.1	20.0	6.8	8.0	8.8	5.5	4.1
Semiconductor Manufacture	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Magnesium Production and Processing	0.0	0.0	+	+	+	+	0.1
PFCs	**24.3**	**6.6**	**3.9**	**4.4**	**6.9**	**6.0**	**5.8**
Aluminum Production	21.5	3.4	1.9	1.9	3.5	2.9	3.0
Semiconductor Manufacture	2.8	3.2	2.0	2.6	3.4	3.0	2.9
SF_6	**31.1**	**14.0**	**9.3**	**9.5**	**10.0**	**7.7**	**6.9**

Electrical Transmission and Distribution	25.4	10.6	7.3	7.0	6.8	5.7	5.1
Magnesium Production and Processing	5.2	2.7	1.6	2.1	2.8	1.6	1.4
Semiconductor Manufacture	0.5	0.7	0.3	0.4	0.4	0.4	0.4
NF_3	**+**	**0.5**	**0.4**	**0.5**	**0.7**	**0.6**	**0.6**
Semiconductor Manufacture	+	0.5	0.4	0.5	0.7	0.6	0.6
Total	**342.1**	**367.4**	**314.9**	**353.6**	**371.0**	**361.2**	**359.1**

Notes: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

Note: Totals may not sum due to independent rounding.

+ Does not exceed 0.05 MMT CO_2 Eq.

[a] Small amounts of PFC emissions also result from this source.

Table 4-2: Emissions from Industrial Processes and Product Use (kt)

Gas/Source	1990	2005	2009	2010	2011	2012	2013
CO_2	**207,166**	**191,101**	**141,126**	**165,737**	**169,727**	**166,359**	**162,979**
Iron and Steel Production & Metallurgical Coke Production	99,781	66,666	43,029	55,746	60,008	54,327	52,288
Iron and Steel Production	*97,311*	*64,623*	*42,073*	*53,662*	*58,583*	*53,786*	*50,466*
Metallurgical Coke Production	*2,470*	*2,043*	*956*	*2,084*	*1,425*	*542*	*1,822*
Cement Production	33,278	45,910	29,432	31,256	32,010	35,051	36,146
Petrochemical Production	21,633	28,124	23,706	27,388	26,396	26,477	26,514
Lime Production	11,700	14,552	11,411	13,381	13,981	13,715	14,072
Ammonia Production	13,047	9,196	8,454	9,188	9,292	9,377	10,152
Urea Consumption for Non-Agricultural Purposes	3,784	3,653	3,427	4,730	4,029	4,449	4,663
Other Process Uses of Carbonates	4,907	6,339	7,583	9,560	9,335	8,022	4,424
Aluminum Production	6,831	4,142	3,009	2,722	3,292	3,439	3,255
Soda Ash Production and Consumption	2,741	2,868	2,488	2,612	2,624	2,672	2,712
Ferroalloy Production	2,152	1,392	1,469	1,663	1,735	1,903	1,785
Titanium Dioxide Production	1,195	1,755	1,648	1,769	1,729	1,528	1,608
Zinc Production	632	1,030	943	1,182	1,286	1,486	1,429
Phosphoric Acid Production	1,586	1,395	1,016	1,130	1,198	1,138	1,173
Glass Production	1,535	1,928	1,045	1,481	1,299	1,248	1,160
Carbon Dioxide Consumption	1,472	1,375	1,795	1,206	802	841	903
Lead Production	516	553	525	542	538	527	525
Silicon Carbide Production and Consumption	375	219	145	181	170	158	169
Magnesium Production and Processing	1	3	1	1	3	2	2
CH_4	**56**	**40**	**20**	**27**	**30**	**33**	**32**
Iron and Steel Production & Metallurgical Coke Production	46	34	17	25	28	29	28
Iron and Steel Production	*46*	*34*	*17*	*25*	*28*	*29*	*28*
Metallurgical Coke Production	+	+	+	+	+	+	+
Petrochemical Production	9	6	2	2	2	3	3
Ferroalloy Production	1	+	+	+	+	1	+
Silicon Carbide Production and Consumption	1	+	+	+	+	+	+
N_2O	**106**	**76**	**56**	**68**	**86**	**69**	**64**
Nitric Acid Production	41	38	32	39	37	35	36
N_2O from Product Uses	14	14	14	14	14	14	14
Adipic Acid Production	51	24	9	14	34	19	13
Semiconductor Manufacture	+	+	+	+	1	1	1
HFCs	**M**	**M**	**M**	**M**	**M**	**M**	**M**

Substitution of Ozone Depleting Substances[a]	M	M	M	M	M	M	M
HCFC-22 Production	3	1	+	1	1	+	+
Semiconductor Manufacture	+	+	+	+	+	+	+
Magnesium Production and Processing	0	0	+	+	+	+	+
PFCs	**M**	**M**	**M**	**M**	**M**	**M**	**M**
Aluminum Production	M	M	M	M	M	M	M
Semiconductor Manufacture	M	M	M	M	M	M	M
SF_6	**2**	**1**	**+**	**+**	**1**	**+**	**+**
Electrical Transmission and Distribution	1	+	+	+	+	+	+
Magnesium Production and Processing	+	+	+	+	+	+	+
Semiconductor Manufacture	+	+	+	+	+	+	+
NF_3	**+**	**+**	**+**	**+**	**+**	**+**	**+**
Semiconductor Manufacture	+	+	+	+	+	+	+

+ Does not exceed 0.5 kt
M (Mixture of gases)
Note: Totals may not sum due to independent rounding.
[a] Small amounts of PFC emissions also result from this source.

The UNFCCC incorporated the *2006 IPCC Guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines)* as the standard for Annex I countries at the Nineteenth Conference of the Parties (Warsaw, November 11-23, 2013). This chapter presents emission estimates calculated in accordance with the methodological guidance provided in these guidelines.

QA/QC and Verification Procedures

For industrial processes and product use sources, a detailed QA/QC plan was developed and implemented. This plan was based on the overall U.S. QA/QC plan, but was tailored to include specific procedures recommended for these sources. Two types of checks were performed using this plan: (1) general, or Tier 1, procedures that focus on annual procedures and checks to be used when gathering, maintaining, handling, documenting, checking, and archiving the data, supporting documents, and files, and (2) source-category specific, or Tier 2, procedures that focus on checks of the emission factors, activity data, and methodologies used for estimating emissions from the relevant industrial process and product use sources. Examples of these procedures include checks to ensure that activity data and emission estimates are consistent with historical trends; that, where possible, consistent and reputable data sources are used across sources; that interpolation or extrapolation techniques are consistent across sources; and that common datasets and factors are used where applicable. Tier 1 quality assurance and quality control procedures have been performed for all industrial process and product use sources. Tier 2 procedures were performed for more significant emission categories, consistent with the IPCC *Good Practice Gudelines*.

For most industrial process and product use categories, activity data is obtained through a survey of manufacturers conducted by various organizations (specified within each source); the uncertainty of the activity data is a function of the reliability of reported plant-level production data and is influenced by the completeness of the survey response. The emission factors used are defaults from IPCC, derived using calculations that assume precise and efficient chemical reactions, or were based upon empirical data in published references. As a result, uncertainties in the emission coefficients can be attributed to, among other things, inefficiencies in the chemical reactions associated with each production process or to the use of empirically-derived emission factors that are biased; therefore, they may not represent U.S. national averages. Additional assumptions are described within each source.

The uncertainty analysis performed to quantify uncertainties associated with the 2013 emission estimates from industrial processes and product use continues a multi-year process for developing credible quantitative uncertainty estimates for these source categories using the IPCC Tier 2 approach. As the process continues, the type and the characteristics of the actual probability density functions underlying the input variables are identified and better characterized (resulting in development of more reliable inputs for the model, including accurate characterization of correlation between variables), based primarily on expert judgment. Accordingly, the quantitative uncertainty estimates reported in this section should be considered illustrative and as iterations of ongoing efforts to produce accurate uncertainty estimates. The correlation among data used for estimating emissions for different sources can influence the uncertainty analysis of each individual source. While the uncertainty analysis recognizes very significant connections among sources, a more comprehensive approach that accounts for all linkages will be identified as the uncertainty analysis moves forward.

Box 4-1: Industrial Processes Data from EPA's Greenhouse Gas Reporting Program

On October 30, 2009, the U.S. EPA published a rule requiring annual of greenhouse gas data from large GHG emissions sources in the United States. Implementation of the rule, codified at 40 CFR part 98, is referred to as EPA's Greenhouse Gas Reporting Program (GHGRP). The rule applies to direct greenhouse gas emitters, fossil fuel suppliers, industrial gas suppliers, and facilities that inject CO_2 underground for sequestration or other reasons and requires reporting by sources or suppliers in 41 industrial categories. Annual reporting is at the facility level, except for certain suppliers of fossil fuels and industrial greenhouse gases. In general, the threshold for reporting is 25,000 metric tons or more of CO_2 Eq. per year, but reporting is required for all facilities in some industries. Calendar year 2010 was the first year for which data were reported for facilities subject to 40 CFR part 98, though some source categories first reported data for calendar year 2011.

EPA's GHGRP dataset and the data presented in this Inventory report are complementary. EPA presents the data collected by EPA's GHGRP through a data publication tool (ghgdata.epa.gov) that allows data to be viewed in several formats, including maps, tables, charts, and graphs for individual facilities or groups of facilities. Most methodologies used in EPA's GHGRP are consistent with IPCC, though for EPA's GHGRP, facilities collect detailed information specific to their operations according to detailed measurement standards. This may differ from the more aggregated data collected for the inventory to estimate total, national U.S. emissions. It should be noted that the definitions for source categories in the GHGRP may differ from those used in this Inventory in meeting the UNFCCC reporting guidelines (IPCC 2011). In line with the UNFCCC reporting guidelines, the Inventory report is a comprehensive accounting of all emissions from source categories identified in the IPCC guidelines. Further information on the reporting categorizations in EPA's GHGRP and specific data caveats associated with monitoring methods in EPA's GHGRP has been provided on the EPA's GHGRP website.

For certain source categories in this Inventory (e.g., nitric acid production and petrochemical production), EPA has also integrated data values that have been calculated by aggregating GHGRP data that is considered confidential business information (CBI) at the facility level. EPA, with industry engagement, has put forth criteria to confirm that a given data aggregation shields underlying CBI from public disclosure. EPA is publishing only data values that meet these aggregation criteria.[149] Specific uses of aggregated facility-level data are described in the respective methodological sections. For other source categories in this chapter, as indicated in the respective planned improvements sections, EPA is continuing to analyze how facility-level GHGRP data may be used to improve the national estimates presented in this Inventory, giving particular consideration to ensuring time series consistency and completeness.

4.1 Cement Production (IPCC Source Category 2A1)

Cement production is an energy- and raw material-intensive process that results in the generation of CO_2 from both the energy consumed in making the cement and the chemical process itself. Emissions from fuels consumed for energy purposes during the production of cement are accounted for in the Energy chapter.

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[149] U.S. EPA Greenhouse Gas Reporting Program, September 16, 2014 Developments on Publication of Aggregated Greenhouse Gas Data, see http://www.epa.gov/climate/ghgreporting/reporters/cbi/index.html

During the cement production process, calcium carbonate ($CaCO_3$) is heated in a cement kiln at a temperature of about 1,450°C (2,400°F) to form lime (i.e., calcium oxide or CaO) and CO_2 in a process known as calcination or calcining. The quantity of CO_2 emitted during cement production is directly proportional to the lime content of the clinker. During calcination, each mole of limestone ($CaCO_3$) heated in the clinker kiln forms one mole of lime (CaO) and one mole of CO_2:

$$CaCO_3 + heat \rightarrow CaO + CO_2$$

Next, the lime is combined with silica-containing materials to produce clinker (an intermediate product), with the earlier byproduct CO_2 being released to the atmosphere. The clinker is then allowed to cool, mixed with a small amount of gypsum and potentially other materials (e.g., slag), and used to make Portland cement.[150]

CO_2 emitted from the chemical process of cement production is the second largest source of industrial CO_2 emissions in the United States. Cement is produced in 35 states and Puerto Rico. Texas, Missouri, California, Pennsylvania, and Florida were the five leading cement-producing States in 2013 and accounted for approximately 48 percent of total U.S. production (USGS 2014). Clinker production in 2013 increased approximately 3 percent from 2012 levels. This increase can be attributed to an increase in spending in new residential construction and nonresidential buildings. In 2013, U.S. clinker production totaled 69,901 kilotons (USGS 2014). The resulting CO_2 emissions were estimated to be 36.1 MMT CO_2 Eq. (36,146 kt) (see Table 4-3).

Table 4-3: CO_2 Emissions from Cement Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	33.3	33,278
2005	45.9	45,910
2009	29.4	29,432
2010	31.3	31,256
2011	32.0	32,010
2012	35.1	35,051
2013	36.1	36,146

Greenhouse gas emissions from cement production increased every year from 1991 through 2006 (with the exception of a slight decrease in 1997), but decreased in the following years until 2009. Emissions from cement production were at their lowest levels in 2009 (2009 emissions are approximately 28 percent lower 2008 emissions and 12 percent lower than 1990). Since 2010, emissions have increased slightly. In 2013, emissions from cement production increased by 3 percent from the 2012 levels.

Emissions since 1990 have increased by 9 percent. Emissions decreased significantly between 2008 and 2009, due to the economic recession and associated decrease in demand for construction materials. Emissions increased slightly from 2009 levels in 2010, and increased slightly again in 2011, 2012, and in 2013 due to increasing consumption. Cement continues to be a critical component of the construction industry; therefore, the availability of public and private construction funding, as well as overall economic conditions, have considerable influence on cement production.

Methodology

CO_2 emissions were estimated using the Tier 2 methodology from the *2006 IPCC Guidelines*. The Tier 2 methodology was used because detailed and complete data (including weights and composition) for carbonate(s) consumed in clinker production are not available, and thus a rigorous Tier 3 approach is impractical. Tier 2 specifies

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[150] Approximately three percent of total clinker production is used to produce masonry cement, which is produced using plasticizers (e.g., ground limestone, lime) and Portland cement (USGS 2011). Carbon dioxide emissions that result from the production of lime used to create masonry cement are included in the Lime Manufacture source category.

the use of aggregated plant or national clinker production data and an emission factor, which is the product of the average lime fraction for clinker of 65 percent and a constant reflecting the mass of CO_2 released per unit of lime. The USGS mineral commodity expert for cement has confirmed that this is a reasonable assumption for the United States (Van Oss 2013a). This calculation yields an emission factor of 0.51 tons of CO_2 per ton of clinker produced, which was determined as follows:

$$\mathrm{EF_{clinker}} = 0.6460\ \mathrm{CaO} \times [(44.01\ \mathrm{g/mole}\ \mathrm{CO_2}) \div (56.08\ \mathrm{g/mole}\ \mathrm{CaO})] = 0.5070\ \mathrm{tons}\ \mathrm{CO_2/ton\ clinker}$$

During clinker production, some of the clinker precursor materials remain in the kiln as non-calcinated, partially calcinated, or fully calcinated cement kiln dust (CKD). The emissions attributable to the calcinated portion of the CKD are not accounted for by the clinker emission factor. The IPCC recommends that these additional CKD CO_2 emissions should be estimated as two percent of the CO_2 emissions calculated from clinker production (when data on CKD generation are not available). Total cement production emissions were calculated by adding the emissions from clinker production to the emissions assigned to CKD (IPCC 2006).

Furthermore, small amounts of impurities (i.e., not calcium carbonate) may exist in the raw limestone used to produce clinker. The proportion of these impurities is generally minimal, although a small amount (1 to 2 percent) of magnesium oxide (MgO) may be desirable as a flux. Per the IPCC Tier 2 methodology, a correction for magnesium oxide is not used, since the amount of magnesium oxide from carbonate is likely very small and the assumption of a 100 percent carbonate source of CaO already yields an overestimation of emissions (IPCC 2006). The 1990 through 2012 activity data for clinker production (see Table 4-4) were obtained from USGS (Van Oss 2013b). Clinker production data for 2013 were also obtained from USGS (USGS 2014).The data were compiled by USGS (to the nearest ton) through questionnaires sent to domestic clinker and cement manufacturing plants, including the facilities in Puerto Rico.

Table 4-4: Clinker Production (kt)

Year	Clinker
1990	64,355
2005	88,783
2009	56,918
2010	60,444
2011	61,903
2012	67,784
2013	69,901

Note: Clinker production from 1990-2013 includes Puerto Rico. Data were obtained from USGS (Van Oss 2013a; USGS 2014), whose original data source was USGS and U.S. Bureau of Mines Minerals Yearbooks (2013 data obtained from mineral industry surveys for cement in June 2014).

Uncertainty and Time-Series Consistency

The uncertainties contained in these estimates are primarily due to uncertainties in the lime content of clinker and in the percentage of CKD recycled inside the cement kiln. Uncertainty is also associated with the assumption that all calcium-containing raw materials are $CaCO_3$, when a small percentage likely consists of other carbonate and non-carbonate raw materials. The lime content of clinker varies from 60 to 67 percent; 65 percent is used as a representative value (Van Oss 2013a). CKD loss can range from 1.5 to 8 percent depending upon plant specifications. Additionally, some amount of CO_2 is reabsorbed when the cement is used for construction. As cement reacts with water, alkaline substances such as calcium hydroxide are formed. During this curing process, these compounds may react with CO_2 in the atmosphere to create calcium carbonate. This reaction only occurs in roughly the outer 0.2 inches of surface area. Because the amount of CO_2 reabsorbed is thought to be minimal, it was not estimated.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-5. Based on the uncertainties associated with total U.S. clinker production, the CO_2 emission factor for clinker production, and the emission factor for additional CO_2 emissions from CKD, 2013 CO_2 emissions from cement production were estimated to be between 34.0 and 38.3 MMT CO_2 Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 6 percent below and 6 percent above the emission estimate of 36.1 MMT CO_2 Eq.

Table 4-5: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Cement Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Cement Production	CO_2	36.1	34.0	38.3	-6%	+6%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Cement Production source category. Particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[151]

4.2 Lime Production (IPCC Source Category 2A2)

Lime is an important manufactured product with many industrial, chemical, and environmental applications. Lime production involves three main processes: stone preparation, calcination, and hydration. Carbon dioxide is generated during the calcination stage, when limestone—mostly calcium carbonate ($CaCO_3$)—is roasted at high temperatures in a kiln to produce CaO and CO_2. The CO_2 is given off as a gas and is normally emitted to the atmosphere.

$$CaCO_3 \rightarrow CaO + CO_2$$

Some of the CO_2 generated during the production process, however, is recovered at some facilities for use in sugar refining and precipitated calcium carbonate (PCC) production.[152] Emissions from fuels consumed for energy purposes during the production of lime are accounted for in the Energy chapter.

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[151] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>

[152] PCC is obtained from the reaction of CO_2 with calcium hydroxide. It is used as a filler and/or coating in the paper, food, and plastic industries.

For U.S. operations, the term “lime” actually refers to a variety of chemical compounds. These include calcium oxide (CaO), or high-calcium quicklime; calcium hydroxide ($Ca(OH)_2$), or hydrated lime; dolomitic quicklime ([CaO•MgO]); and dolomitic hydrate ([$Ca(OH)_2$•MgO] or [$Ca(OH)_2$•$Mg(OH)_2$]).

The contemporary lime market is approximately distributed across five end-use categories as follows: metallurgical uses, 38 percent; environmental uses, 31 percent; chemical and industrial uses, 22 percent; construction uses, 8 percent; and refractory dolomite, 1 percent. The major uses are in steel making, flue gas desulfurization systems at coal-fired electric power plants, construction, and water purification. Lime is also used as a CO_2 scrubber, and there has been experimentation on the use of lime to capture CO_2 from electric power plants.

Lime production in the United States—including Puerto Rico— was reported to be 19,210 kilotons in 2013 (Corathers 2014). Principal lime producing states are Alabama, Kentucky, Missouri, Nevada, Ohio, Pennsylvania, and Texas.

U.S. lime production resulted in estimated net CO_2 emissions of 14.1 MMT CO_2 Eq. (14,072 kt) (see Table 4-6 and Table 4-7). The trends in CO_2 emissions from lime production are directly proportional to trends in production, which are described below.

Table 4-6: CO_2 Emissions from Lime Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	11.7	11,700
2005	14.6	14,552
2009	11.4	11,411
2010	13.4	13,381
2011	14.0	13,981
2012	13.7	13,715
2013	14.1	14,072

Table 4-7: Potential, Recovered, and Net CO_2 Emissions from Lime Production (kt)

Year	Potential	Recovered[a]	Net Emissions
1990	11,959	259	11,700
2005	15,074	522	14,552
2009	11,872	461	11,411
2010	13,776	395	13,381
2011	14,389	407	13,981
2012	14,188	473	13,715
2013	14,539	467	14,072

[a] For sugar refining and PCC production.

Note: Totals may not sum due to independent rounding.

In 2013, lime production was nearly the same as 2011 levels (increase of 1 percent) at 19,210 kilotons. In 2013, lime production increased from 2012 levels by approximately 3 percent. Lime production in 2010 rebounded from a 21 percent decline in 2009 to 18,219 kilotons, which is still 8 percent below 2008 levels. Lime production declined in 2009 mostly due to the economic recession and the associated significant downturn in major markets such as construction and steel. The surprising rebound in 2010 is primarily due to increased consumption in steelmaking, chemical and industrial uses, and in flue gas desulfurization.

Methodology

To calculate emissions, the amounts of high-calcium and dolomitic lime produced were multiplied by their respective emission factors using the Tier 2 approach from the *2006 IPCC Guidelines* (IPCC 2006). The emission factor is the product of the stoichiometric ratio between CO_2 and CaO, and the average CaO and MgO content for lime. The CaO and MgO content for lime is assumed to be 95 percent for both high-calcium and dolomitic lime) (IPCC 2006). The emission factors were calculated as follows:

For high-calcium lime:

$$[(44.01\ \mathrm{g/mole\ CO_2}) \div (56.08\ \mathrm{g/mole\ CaO})] \times (0.9500\ \mathrm{CaO/lime}) = 0.7455\ \mathrm{g\ CO_2/g\ lime}$$

For dolomitic lime:

$$[(88.02\ \mathrm{g/mole\ CO_2}) \div (96.39\ \mathrm{g/mole\ CaO})] \times (0.9500\ \mathrm{CaO/lime}) = 0.8675\ \mathrm{g\ CO_2/g\ lime}$$

Production was adjusted to remove the mass of chemically combined water found in hydrated lime, determined according to the molecular weight ratios of H_2O to ($Ca(OH)_2$ and [$Ca(OH)_2$•$Mg(OH)_2$]) (IPCC 2006). These factors set the chemically combined water content to 24.3 percent for high-calcium hydrated lime, and 27.2 percent for dolomitic hydrated lime.

The *2006 IPCC Guidelines* (Tier 2 method) also recommends accounting for emissions from lime kiln dust (LKD). LKD is a byproduct of the lime manufacturing process. LKD is a very fine-grained material and is especially useful for applications requiring very small particle size. Most common LKD applications include soil reclamation and agriculture. Currently, data on annual LKD production is not readily available. Lime emission estimates were multiplied by a factor of 1.02 to account for emissions from LKD (IPCC 2006).

Lime emission estimates were further adjusted to account for the amount of CO_2 captured for use in on-site processes. All the domestic lime facilities are required to report these data to EPA under its GHGRP. The total national-level annual amount of CO_2 captured for on-site process use was obtained from EPA's GHGRP (EPA 2014) based on reported facility level data. The amount of CO_2 captured/recovered for on-site process use is deducted from the total potential emissions (i.e., from lime production and LKD). The net lime emissions are presented in Table 4-6 and Table 4-7. GHGRP data on CO_2 removals (i.e., CO_2 captured/recovered) was available only for 2010 through 2013. Since GHGRP data are not available for 1990 through 2009, IPCC "splicing" techniques were used as per the *2006 IPCC Guidelines* on time series consistency (*2006 IPCC Guidelines*, Volume 1, Chapter 5). The prior estimates for CO_2 removal for 1990 through 2009 were adjusted based on the "overlap" technique recommended by IPCC. Refer to the Recalculations Discussion section, below, for more details.

Lime production data (by type, high-calcium- and dolomitic-quicklime, high-calcium- and dolomitic-hydrated, and dead-burned dolomite) for 1990 through 2013 (see Table 4-8) were obtained from USGS (1992 through 2013, Corathers 2014) and are compiled by USGS to the nearest ton. Natural hydraulic lime, which is produced from CaO and hydraulic calcium silicates, is not manufactured in the United States (USGS 2011). Total lime production was adjusted to account for the water content of hydrated lime by converting hydrate to oxide equivalent based on recommendations from the IPCC, and is presented in Table 4-9 (IPCC 2006). The CaO and CaO•MgO contents of lime were obtained from the IPCC (IPCC 2006). Since data for the individual lime types (high calcium and dolomitic) were not provided prior to 1997, total lime production for 1990 through 1996 was calculated according to the three year distribution from 1997 to 1999.

Table 4-8: High-Calcium- and Dolomitic-Quicklime, High-Calcium- and Dolomitic-Hydrated, and Dead-Burned-Dolomite Lime Production (kt)

Year	High-Calcium Quicklime	Dolomitic Quicklime	High-Calcium Hydrated	Dolomitic Hydrated	Dead-Burned Dolomite
1990	11,166	2,234	1,781	319	342
2005	14,100	2,990	2,220	474	200
2009	11,800	1,830	1,690	261	200
2010	13,300	2,570	1,910	239	200

Year	High-Calcium Quicklime	Dolomitic Quicklime	High-Calcium Hydrated	Dolomitic Hydrated	Dead-Burned Dolomite
2011	13,900	2,690	2,010	230	200
2012	13,600	2,710	2,020	237	200
2013	13,800	2,870	2,050	260	230

Table 4-9: Adjusted Lime Production (kt)

Year	High-Calcium	Dolomitic
1990	12,466	2,800
2005	15,721	3,522
2009	13,034	2,213
2010	14,694	2,937
2011	15,367	3,051
2012	15,075	3,076
2013	15,297	3,282

Note: Minus water content of hydrated lime

Uncertainty and Time Series Consistency

The uncertainties contained in these estimates can be attributed to slight differences in the chemical composition of lime products and CO_2 recovery rates for on-site process use over the time series. Although the methodology accounts for various formulations of lime, it does not account for the trace impurities found in lime, such as iron oxide, alumina, and silica. Due to differences in the limestone used as a raw material, a rigid specification of lime material is impossible. As a result, few plants produce lime with exactly the same properties.

In addition, a portion of the CO_2 emitted during lime production will actually be reabsorbed when the lime is consumed, especially at captive lime production facilities. As noted above, lime has many different chemical, industrial, environmental, and construction applications. In many processes, CO_2 reacts with the lime to create calcium carbonate (e.g., water softening). Carbon dioxide reabsorption rates vary, however, depending on the application. For example, 100 percent of the lime used to produce precipitated calcium carbonate reacts with CO_2; whereas most of the lime used in steel making reacts with impurities such as silica, sulfur, and aluminum compounds. Quantifying the amount of CO_2 that is reabsorbed would require a detailed accounting of lime use in the United States and additional information about the associated processes where both the lime and byproduct CO_2 are "reused" are required to quantify the amount of CO_2 that is reabsorbed. Research conducted thus far has not yielded the necessary information to quantify CO_2 reabsorption rates.[153] However, some additional information on the amount of CO_2 consumed on site at lime facilities has been obtained from EPA's GHGRP.

In some cases, lime is generated from calcium carbonate byproducts at pulp mills and water treatment plants.[154] The lime generated by these processes is included in the USGS data for commercial lime consumption. In the pulping industry, mostly using the Kraft (sulfate) pulping process, lime is consumed in order to causticize a process liquor (green liquor) composed of sodium carbonate and sodium sulfide. The green liquor results from the dilution of the smelt created by combustion of the black liquor where biogenic C is present from the wood. Kraft mills

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[153] Representatives of the National Lime Association estimate that CO_2 reabsorption that occurs from the use of lime may offset as much as a quarter of the CO_2 emissions from calcination (Males 2003).

[154] Some carbide producers may also regenerate lime from their calcium hydroxide byproducts, which does not result in emissions of CO_2. In making calcium carbide, quicklime is mixed with coke and heated in electric furnaces. The regeneration of lime in this process is done using a waste calcium hydroxide (hydrated lime) [$CaC_2 + 2H_2O \rightarrow C_2H_2 + Ca(OH)_2$], not calcium carbonate [$CaCO_3$]. Thus, the calcium hydroxide is heated in the kiln to simply expel the water [$Ca(OH)_2 + heat \rightarrow CaO + H_2O$] and no CO_2 is released.

recover the calcium carbonate “mud” after the causticizing operation and calcine it back into lime—thereby generating CO_2—for reuse in the pulping process. Although this re-generation of lime could be considered a lime manufacturing process, the CO_2 emitted during this process is mostly biogenic in origin, and therefore is not included in the industrial processes totals (Miner and Upton 2002). In accordance with IPCC methodological guidelines, any such emissions are calculated by accounting for net carbon (C) fluxes from changes in biogenic C reservoirs in wooded or crop lands (see the *Land Use, Land-Use Change, and Forestry* chapter).

In the case of water treatment plants, lime is used in the softening process. Some large water treatment plants may recover their waste calcium carbonate and calcine it into quicklime for reuse in the softening process. Further research is necessary to determine the degree to which lime recycling is practiced by water treatment plants in the United States.

Another uncertainty is the assumption that calcination emissions for LKD are around 2 percent. The National Lime association has commented that the estimates of emissions from LKD in the United States could be closer to 6 percent. They also note that additional emissions (~2 percent) may also be generated through production of other byproducts/wastes (off-spec lime that is not recycled, scrubber sludge) at lime plants (Seeger 2013). There is limited data publicly available on LKD generation rates and also quantities, types of other byproducts/wastes produced at lime facilities. Further research and data is needed to improve understanding of additional calcination emissions to consider revising the current assumptions that are based on the IPCC Guidelines.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-10. Lime CO_2 emissions for 2013 were estimated to be between 13.7 and 14.4 MMT CO_2 Eq. at the 95 percent confidence level. This confidence level indicates a range of approximately 3 percent below and 3 percent above the emission estimate of 14.1 MMT CO_2 Eq.

Table 4-10: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Lime Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Lime Production	CO_2	14.1	13.7	14.4	-3%	+3%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Updated CO_2 recovery data was used for this category, aggregating reported facility level data from the GHGRP data on amount of CO_2 captured for on-site process use 2010 through 2013 (EPA 2014). Since these data were not available for the entire time series, IPCC-recommended “splicing” techniques were followed to estimate CO_2 removals for 1990 through 2009. In cases where the same method and data source is not available for the entire time series, IPCC recommends the use of “splicing” techniques to maintain time series consistency.

Of these, overlap is the only suitable method that could be applied to revise the 1990 through 2009 CO_2 removal estimates. The surrogate data method is not applicable due to absence of appropriate surrogate data for CO_2 removal. Interpolation and trend extrapolation methods are not suitable for longer time-periods (1990 through 2009). Therefore, the overlap method was selected to revise the prior 1990 through 2009 removal estimates.

According to the IPCC overlap method (IPCC 2006), the prior CO_2 removal estimates for 1990 through 2009 were multiplied by an adjustment factor. The adjustment factor is the average ratio of the removal estimates prepared using the new and the method previously used during the period of overlap (2010 through 2013).

$$y_0 = x_0 \times \left(\frac{1}{(n-m+1)} \times \sum_{i=m}^{n} \frac{y_i}{x_i} \right)$$

where:

y_0 = the recalculated emission or removal estimate computed using overlap method
x_0 = the estimate developed using the previously used method
y_i = estimate(s) prepared using the new method during the period of overlap (2010-2013)
x_i = estimate(s) prepared using the previously-used method during the period of overlap (2010-2013)
m = starting year for the period of overlap (2010)
n = ending year for the period of overlap (2013)

Using the above equation, the adjustment factor was calculated to be 0.4815. The prior removal estimates for 1990 through 2009 were multiplied by this adjustment factor to obtain the revised removal estimates. This change resulted in a decrease of the annual CO_2 removal estimates by approximately half. As a result of the decreased removal estimates, the net CO_2 emissions from lime production increase for the entire time series. In the previous Inventory reports, the CO_2 removal estimates (i.e., CO_2 captured/recovered) were calculated using lime consumption data for PCC production and sugar refining. PCC producers and sugar refineries recover CO_2 emitted by lime production facilities for use as an input into production or refining processes. For CO_2 recovery by sugar refineries, lime consumption estimates (Corathers 2014) were multiplied by a CO_2 recovery factor to determine the total amount of CO_2 recovered from lime production facilities. According to industry outreach by state agencies and USGS, sugar refineries use captured CO_2 for 100 percent of their CO_2 input (Lutter 2009, Miller 2013). Carbon dioxide recovery by PCC producers was determined by multiplying lime consumption for PCC production (USGS 1992 through 2013, Corathers 2014) with the percentage CO_2 of production weight for PCC production at lime plants (i.e., $CO_2/CaCO_3$ = 44/100) and a CO_2 recovery factor based on the amount of purchased CO_2 by PCC manufacturers (Prillaman 2008 through 2012, Miller 2013). As data were only available starting in 2007, CO_2 recovery for the period 1990 through 2006 was extrapolated by determining a ratio of PCC production at lime facilities to lime consumption for PCC (USGS 1992 through 2008).

Planned Improvements

Future improvements involve continuing research to improve current assumptions associated with emissions from production of LKD and other byproducts/wastes as discussed in the Uncertainty and Time Series Consistency section per comments from the National Lime Association. Pending resources and data availability, historical CO_2 recovery rates at U.S. facilities producing lime will be investigated to further evaluate results from use of overlap method to improve time series consistency.

4.3 Glass Production (IPCC Source Category 2A3)

Glass production is an energy and raw-material intensive process that results in the generation of CO_2 from both the energy consumed in making glass and the glass process itself. Emissions from fuels consumed for energy purposes during the production of glass are accounted for in the Energy sector.

Glass production employs a variety of raw materials in a glass-batch. These include formers, fluxes, stabilizers, and sometimes colorants. The major raw materials (i.e., fluxes and stabilizers) which emit process-related CO_2 emissions during the glass melting process are limestone, dolomite, and soda ash. The main former in all types of glass is silica (SiO_2). Other major formers in glass include feldspar and boric acid (i.e., borax). Fluxes are added to lower the temperature at which the batch melts. Most commonly used flux materials are soda ash (sodium carbonate, Na_2CO_3) and potash (potassium carbonate, K_2O). Stabilizers are used to make glass more chemically stable and to keep the finished glass from dissolving and/or falling apart. Commonly used stabilizing agents in glass production are limestone ($CaCO_3$), dolomite ($CaCO_3MgCO_3$), alumina (Al_2O_3), magnesia (MgO), barium carbonate ($BaCO_3$), strontium carbonate ($SrCO_3$), lithium carbonate (Li_2CO_3), and zirconia (ZrO_2) (OIT 2002). Glass makers also use a certain amount of recycled scrap glass (cullet), which comes from in-house return of glassware broken in the process or other glass spillage or retention such as recycling or cullet broker services.

The raw materials (primarily limestone, dolomite and soda ash) release CO_2 emissions in a complex high-temperature chemical reaction during the glass melting process. This process is not directly comparable to the calcination process used in lime manufacturing, cement manufacturing, and Process Carbonates Use (i.e., limestone/dolomite use), but has the same net effect in terms of CO_2 emissions (IPCC 2006). The U.S. glass industry can be divided into four main categories: containers, flat (window) glass, fiber glass, and specialty glass. The majority of commercial glass produced is container and flat glass (EPA 2009). The United States is one of the major global exporters of glass. Domestically, demand comes mainly from the construction, auto, bottling, and container industries. There are over 1,500 companies that manufacture glass in the United States, with the largest being Corning, Guardian Industries, Owens-Illinois, and PPG Industries.[155]

In 2013, 335 kilotons of limestone and 2,440 kilotons of soda ash were consumed for glass production in 2013 (USGS 2014b, Willett 2014). Dolomite consumption data for glass manufacturing was not publicly available for 2013. Use of limestone and soda ash in glass production resulted in aggregate CO_2 emissions of 1.2 MMT CO_2 Eq. (1,160 kt) (see Table 4-11). Overall, emissions have decreased 24 percent from 1990 through 2013.

Emissions from glass production have remained relatively constant over the time series with some fluctuations since 1990. In general, these fluctuations were related to the behavior of the export market and the U.S. economy. Specifically, the extended downturn in residential and commercial construction and automotive industries between 2008 and 2010 resulted in reduced consumption of glass products, causing a drop in global demand for limestone/dolomite and soda ash, and a corresponding decrease in emissions. Furthermore, the glass container sector is one of the leading soda ash consuming sectors in the United States. Some commercial food and beverage package manufacturers are shifting from glass containers towards lighter and more cost effective polyethylene terephthalate (PET) based containers, putting downward pressure on domestic consumption of soda ash (USGS 1995 through 2013b).

Table 4-11: CO_2 Emissions from Glass Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.5	1,535
2005	1.9	1,928
2009	1.0	1,045
2010	1.5	1,481
2011	1.3	1,299
2012	1.2	1,248
2013	1.2	1,160

Methodology

CO_2 emissions were calculated based on the *IPCC 2006 Guidelines* Tier 3 method by multiplying the quantity of input carbonates (limestone, dolomite, and soda ash) by the carbonate-based emission factor (in metric tons CO_2/metric ton carbonate): limestone, 0.43971; dolomite, 0.47732; and soda ash, 0.41492.

Consumption data for 1990 through 2013 of limestone, dolomite, and soda ash used for glass manufacturing were obtained from the USGS *Minerals Yearbook: Crushed Stone Annual Report* (1995 through 2014), 2013 preliminary data from the USGS Crushed Stone Commodity Expert (Willett 2014), the *USGS Minerals Yearbook: Soda Ash Annual Report* (1995 through 2013), USGS Mineral Industry Surveys for Soda Ash in August 2014 (USGS 2014)

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[155] Excerpt from Glass & Glass Product Manufacturing Industry Profile, First Research. Available online at <http://www.firstresearch.com/Industry-Research/Glass-and-Glass-Product-Manufacturing.html>.

and the U.S. Bureau of Mines (1991 and 1993a), which are reported to the nearest ton. During 1990 and 1992, the USGS did not conduct a detailed survey of limestone and dolomite consumption by end-use. Consumption for 1990 was estimated by applying the 1991 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to 1990 total use. Similarly, the 1992 consumption figures were approximated by applying an average of the 1991 and 1993 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to the 1992 total.

Additionally, each year the USGS withholds data on certain limestone and dolomite end-uses due to confidentiality agreements regarding company proprietary data. For the purposes of this analysis, emissive end-uses that contained withheld data were estimated using one of the following techniques: (1) the value for all the withheld data points for limestone or dolomite use was distributed evenly to all withheld end-uses; or (2) the average percent of total limestone or dolomite for the withheld end-use in the preceding and succeeding years.

There is a large quantity of limestone and dolomite reported to the USGS under the categories "unspecified–reported" and "unspecified–estimated." A portion of this consumption is believed to be limestone or dolomite used for glass manufacturing. The quantities listed under the "unspecified" categories were, therefore, allocated to glass manufacturing according to the percent limestone or dolomite consumption for glass manufacturing end use for that year.[156]

Based on the 2013 reported data, the estimated distribution of soda ash consumption for glass production compared to total domestic soda ash consumption is 48 percent (USGS 2014b).

Table 4-12: Limestone, Dolomite, and Soda Ash Consumption Used in Glass Production (kt)

Activity	**1990**	**2005**	**2009**	**2010**	**2011**	**2012**	**2013**
Limestone	430	920	139	999	614	555	335
Dolomite	59	541	0	0	0	0	0
Soda Ash	3,177	3,050	2,370	2,510	2,480	2,420	2,440
Total	**3,666**	**4,511**	**2,509**	**3,509**	**3,094**	**2,975**	**2,775**

Uncertainty and Time-Series Consistency

The uncertainty levels presented in this section arise in part due to variations in the chemical composition of limestone used in glass production. In addition to calcium carbonate, limestone may contain smaller amounts of magnesia, silica, and sulfur, among other minerals (potassium carbonate, strontium carbonate and barium carbonate, and dead burned dolomite). Similarly, the quality of the limestone (and mix of carbonates) used for glass manufacturing will depend on the type of glass being manufactured.

The estimates below also account for uncertainty associated with activity data. Large fluctuations in reported consumption exist, reflecting year-to-year changes in the number of survey responders. The uncertainty resulting from a shifting survey population is exacerbated by the gaps in the time series of reports. The accuracy of distribution by end use is also uncertain because this value is reported by the manufacturer of the input carbonates (limestone, dolomite & soda ash) and not the end user. For 2013, there has been no reported consumption of dolomite for glass manufacturing. This data has been reported to USGS by dolomite manufacturers and not end-users (i.e., glass manufacturers). There is a high uncertainty associated with this estimate, as dolomite is a major raw material consumed in glass production. Additionally, there is significant inherent uncertainty associated with estimating withheld data points for specific end uses of limestone and dolomite. The uncertainty of the estimates for limestone and dolomite used in glass making is especially high; however, since glass making accounts for a small percent of consumption, its contribution to the overall emissions estimate is low. Lastly, much of the limestone consumed in the United States is reported as "other unspecified uses;" therefore, it is difficult to accurately allocate this unspecified quantity to the correct end-uses. Further research is needed into alternate and more complete sources of data on carbonate-based raw material consumption by the glass industry.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-13. In 2013, glass production CO_2 emissions were estimated to be between 1.1 and 1.2 MMT CO_2 Eq. at the 95 percent confidence

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[156] This approach was recommended by USGS.

level. This indicates a range of approximately 5 percent below and 5 percent above the emission estimate of 1.2 MMT CO_2 Eq.

Table 4-13: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Glass Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Glass Production	CO_2	1.2	1.1	1.2	-5%	+5%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Limestone consumption data for 2012 were revised to reflect updated USGS data. This change resulted in an insignificant increase of CO_2 emissions (less than 1 kt of CO_2). The preliminary data for 2012 was obtained directly from the USGS Crushed Stone Commodity Expert (Willett 2013). In June 2014, USGS published the 2012 Minerals Yearbook for Crushed Stone and the preliminary data was revised to reflect the latest USGS published data. The published time series was reviewed to ensure time series consistency. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Currently, only limestone and soda ash consumption data for glass manufacturing is publicly available. While limestone and soda ash are the predominant carbonates used in glass manufacturing, there are other carbonates that are also consumed for glass manufacturing, although in smaller quantities (e.g. dolomite). Pending resources, future improvements will include research into other sources of data for carbonate consumption by the glass industry.

Additionally, future improvements will also involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Glass Production source category. Particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. Further, EPA's GHGRP has an emission threshold for reporting, so the data do not account for all glass production in the United States. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[157]

4.4 Other Process Uses of Carbonates (IPCC Source Category 2A4)

Limestone ($CaCO_3$), dolomite ($CaCO_3MgCO_3$)[158], and other carbonates such as magnesium carbonate and iron carbonate are basic materials used by a wide variety of industries, including construction, agriculture, chemical,

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[157] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

[158] Limestone and dolomite are collectively referred to as limestone by the industry, and intermediate varieties are seldom distinguished.

metallurgy, glass production, and environmental pollution control. This section addresses only limestone and dolomite use. For industrial applications, carbonates such as limestone and dolomite are heated sufficiently enough to calcine the material and generate CO_2 as a byproduct.

$$CaCO_3 \rightarrow CaO + CO_2$$

$$MgCO_3 \rightarrow MgO + CO_2$$

Examples of such applications include limestone used as a flux or purifier in metallurgical furnaces, as a sorbent in flue gas desulfurization (FGD) systems for utility and industrial plants, and as a raw material for the production of glass, lime, and cement. Emissions from limestone and dolomite used in other process sectors such as cement, lime, glass production, and iron and steel, are excluded from this section and reported under their respective source categories (e.g., glass manufacturing IPCC Source Category 2A7.) Emissions from fuels consumed for energy purposes during these processes are accounted for in the Energy chapter.

Limestone is widely distributed throughout the world in deposits of varying sizes and degrees of purity. Large deposits of limestone occur in nearly every state in the United States, and significant quantities are extracted for industrial applications. The leading limestone producing States are Texas, Missouri, Pennsylvania, Kentucky, and Ohio (USGS 2014). Similarly, dolomite deposits are also widespread throughout the world. Dolomite deposits are found in the United States, Canada, Mexico, Europe, Africa, and Brazil. In the United States, the leading dolomite producing states are Illinois, Pennsylvania, New York, Michigan, and Indiana (USGS 2013c).

In 2013, 10,010 kt of limestone and 1,212 kt of dolomite were consumed for these emissive applications, excluding glass manufacturing (Willett 2014). Usage of limestone and dolomite resulted in aggregate CO_2 emissions of 4.4 MMT CO_2 Eq. (4,424 kt) (see Table 4-14 and Table 4-15). Overall, emissions have decreased 10 percent from 1990 through 2013.

Table 4-14: CO_2 Emissions from Other Process Uses of Carbonates (MMT CO_2 Eq.)

Year	Flux Stone	FGD	Magnesium Production	Other Miscellaneous Uses	Total
1990	2.6	1.4	0.1	0.8	4.9
2005	2.6	3.0	+	0.7	6.3
2009	1.8	5.4	+	0.4	7.6
2010	1.6	7.1	+	0.9	9.6
2011	1.5	5.4	+	2.4	9.3
2012	1.1	5.8	+	1.1	8.0
2013	0.9	3.0	+	0.5	4.4

Notes: Totals may not sum due to independent rounding. “Other miscellaneous uses” include chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining.

+ Emissions are less than 0.05 MMT CO_2 Eq.

Table 4-15: CO_2 Emissions from Other Process Uses of Carbonates (kt)

Year	Flux Stone	FGD	Magnesium Production	Other Miscellaneous Uses	Total
1990	2,592	1,432	64	819	4,907
2005	2,649	2,973	+	718	6,339
2009	1,784	5,403	+	396	7,583
2010	1,560	7,064	+	937	9,560
2011	1,467	5,420	+	2,449	9,335
2012	1,077	5,797	+	1,148	8,022
2013	947	3,002	+	474	4,424

+ Emissions are less than 0.5 kt

Methodology

CO_2 emissions were calculated based on the *2006 IPCC Guidelines* Tier 2 method by multiplying the quantity of limestone or dolomite consumed by the emission factor for limestone or dolomite calcination, respectively, Table 2.1–limestone: 0.43971 tonne CO_2/tonne carbonate, and dolomite: 0.47732 tonne CO_2/tonne carbonate.[159] This methodology was used for flux stone, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining. Flux stone used during the production of iron and steel was deducted from the Other Process Uses of Carbonates estimate and attributed to the Iron and Steel Production estimate. Similarly limestone and dolomite consumption for glass manufacturing, cement, and lime manufacturing are excluded from this category and attributed to their respective categories.

Historically, the production of magnesium metal was the only other significant use of limestone and dolomite that produced CO_2 emissions. At the end of 2001, the sole magnesium production plant operating in the United States that produced magnesium metal using a dolomitic process that resulted in the release of CO_2 emissions ceased its operations (USGS 1995 through 2012b; USGS 2013a).

Consumption data for 1990 through 2013 of limestone and dolomite used for flux stone, flue gas desulfurization systems, chemical stone, mine dusting or acid water treatment, acid neutralization, and sugar refining (see Table 4-16) were obtained from the USGS *Minerals Yearbook: Crushed Stone Annual Report* (1995 through 2014), preliminary data for 2013 from USGS Crushed Stone Commodity Expert (Willett, 2014), and the U.S. Bureau of Mines (1991 and 1993a), which are reported to the nearest ton. The production capacity data for 1990 through 2013 of dolomitic magnesium metal also came from the USGS (1995 through 2012, USGS 2013a) and the U.S. Bureau of Mines (1990 through 1993b). During 1990 and 1992, the USGS did not conduct a detailed survey of limestone and dolomite consumption by end-use. Consumption for 1990 was estimated by applying the 1991 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to 1990 total use. Similarly, the 1992 consumption figures were approximated by applying an average of the 1991 and 1993 percentages of total limestone and dolomite use constituted by the individual limestone and dolomite uses to the 1992 total.

Additionally, each year the USGS withholds data on certain limestone and dolomite end-uses due to confidentiality agreements regarding company proprietary data. For the purposes of this analysis, emissive end-uses that contained withheld data were estimated using one of the following techniques: (1) the value for all the withheld data points for limestone or dolomite use was distributed evenly to all withheld end-uses; (2) the average percent of total limestone or dolomite for the withheld end-use in the preceding and succeeding years; or (3) the average fraction of total limestone or dolomite for the end-use over the entire time period.

There is a large quantity of crushed stone reported to the USGS under the category “unspecified uses.” A portion of this consumption is believed to be limestone or dolomite used for emissive end uses. The quantity listed for “unspecified uses” was, therefore, allocated to each reported end-use according to each end-use’s fraction of total consumption in that year.[160]

Table 4-16: Limestone and Dolomite Consumption (kt)

Activity	1990		2005		2009	2010	2011	2012	2013
Flux Stone	6,737		7,022		4,623	4,440	4,396	3,666	3,317
Limestone	5,804		3,165		1,631	1,921	2,531	3,108	2,119
Dolomite	933		3,857		2,992	2,520	1,865	559	1,199
FGD	3,258		6,761		12,288	16,064	12,326	13,185	6,827
Other Miscellaneous Uses	1,835		1,632		898	2,121	5,548	2,610	1,078
Total	**11,830**		**15,415**		**17,809**	**22,626**	**22,270**	**19,461**	**11,222**

Uncertainty and Time-Series Consistency

The uncertainty levels presented in this section account for uncertainty associated with activity data. Data on limestone and dolomite consumption are collected by USGS through voluntary national surveys. USGS contacts the

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[159] *IPCC 2006 Guidelines*, Volume 3: Chapter 2

[160] This approach was recommended by USGS, the data collection agency.

mines (i.e., producers of various types of crushed stone) for annual sales data. Data on other carbonate consumption are not readily available. The producers report the annual quantity sold to various end-users/industry types. USGS estimates the historical response rate for the crushed stone survey to be approximately 70 percent, the rest is estimated by USGS. Large fluctuations in reported consumption exist, reflecting year-to-year changes in the number of survey responders. The uncertainty resulting from a shifting survey population is exacerbated by the gaps in the time series of reports. The accuracy of distribution by end use is also uncertain because this value is reported by the producer/mines and not the end user. Additionally, there is significant inherent uncertainty associated with estimating withheld data points for specific end uses of limestone and dolomite. Lastly, much of the limestone consumed in the United States is reported as "other unspecified uses;" therefore, it is difficult to accurately allocate this unspecified quantity to the correct end-uses.

Uncertainty in the estimates also arises in part due to variations in the chemical composition of limestone. In addition to calcium carbonate, limestone may contain smaller amounts of magnesia, silica, and sulfur, among other minerals. The exact specifications for limestone or dolomite used as flux stone vary with the pyrometallurgical process and the kind of ore processed.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-17. Other Process Uses of Carbonates CO_2 emissions in 2013 were estimated to be between 4.1 and 4.8 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 8 percent below and 8 percent above the emission estimate of 4.4 MMT CO_2 Eq.

Table 4-17: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Other Process Uses of Carbonates (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Other Process Uses of Carbonates	CO_2	4.4	4.1	4.8	-8%	+8%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

4.5 Ammonia Production (IPCC Source Category 2B1)

Emissions of CO_2 occur during the production of synthetic ammonia, primarily through the use of natural gas, petroleum coke, or naphtha as a feedstock. The natural gas-, naphtha-, and petroleum coke-based processes produce CO_2 and hydrogen (H_2), the latter of which is used in the production of ammonia. Emissions from fuels consumed for energy purposes during the production of ammonia are accounted for in the Energy chapter.

In the United States, the majority of ammonia is produced using a natural gas feedstock; however one synthetic ammonia production plant located in Kansas is producing ammonia from petroleum coke feedstock. In some U.S. plants, some of the CO_2 produced by the process is captured and used to produce urea rather than being emitted to the atmosphere. There are approximately 13 companies operating 25 ammonia producing facilities in 16 states. More than 57 percent of domestic ammonia production capacity is concentrated in the States of Louisiana (30 percent), Oklahoma (21 percent), and Texas (6 percent) (USGS 2014). The brine electrolysis process for production of ammonia does not lead to process-based CO_2 emissions.

There are five principal process steps in synthetic ammonia production from natural gas feedstock. The primary reforming step converts CH_4 to CO_2, carbon monoxide (CO), and H_2 in the presence of a catalyst. Only 30 to 40 percent of the CH_4 feedstock to the primary reformer is converted to CO and CO_2 in this step of the process. The secondary reforming step converts the remaining CH_4 feedstock to CO and CO_2. The CO in the process gas from the secondary reforming step (representing approximately 15 percent of the process gas) is converted to CO_2 in the presence of a catalyst, water, and air in the shift conversion step. Carbon dioxide is removed from the process gas by the shift conversion process, and the hydrogen gas is combined with the nitrogen (N_2) gas in the process gas during the ammonia synthesis step to produce ammonia. The CO_2 is included in a waste gas stream with other process impurities and is absorbed by a scrubber solution. In regenerating the scrubber solution, CO_2 is released from the solution.

The conversion process for conventional steam reforming of CH_4, including the primary and secondary reforming and the shift conversion processes, is approximately as follows:

$$0.88CH_4 + 1.26Air + 1.24H_2O \rightarrow 0.88CO_2 + N_2 + 3H_2$$

$$N_2 + 3H_2 \rightarrow 2NH_3$$

To produce synthetic ammonia from petroleum coke, the petroleum coke is gasified and converted to CO_2 and H_2. These gases are separated, and the H_2 is used as a feedstock to the ammonia production process, where it is reacted with N_2 to form ammonia.

Not all of the CO_2 produced during the production of ammonia is emitted directly to the atmosphere. Some of the ammonia and some of the CO_2 produced by the synthetic ammonia process are used as raw materials in the production of urea [$CO(NH_2)_2$], which has a variety of agricultural and industrial applications.

The chemical reaction that produces urea is:

$$2NH_3 + CO_2 \rightarrow NH_2COONH_4 \rightarrow CO(NH_2)_2 + H_2O$$

Only the CO_2 emitted directly to the atmosphere from the synthetic ammonia production process are accounted for in determining emissions from ammonia production. The CO_2 that is captured during the ammonia production process and used to produce urea does not contribute to the CO_2 emission estimates for ammonia production presented in this section. Instead, CO_2 emissions resulting from the consumption of urea are attributed to the urea consumption or urea application source category (under the assumption that the carbon stored in the urea during its manufacture is released into the environment during its consumption or application). Emissions of CO_2 resulting from agricultural applications of urea are accounted for in the Cropland Remaining Cropland section of the *Land Use, Land-Use Change, and Forestry* chapter. Emissions of CO_2 resulting from non-agricultural applications of urea (e.g., use as a feedstock in chemical production processes) are accounted for in the Urea Consumption for Non-Agricultural Purposes section of this chapter.

Total emissions of CO_2 from ammonia production in 2013 were 10.2 MMT CO_2 Eq. (10,152 kt), and are summarized in Table 4-18 and Table 4-19. Ammonia production relies on natural gas as both a feedstock and a fuel, and as such, market fluctuations and volatility in natural gas prices affect the production of ammonia. Since 1990, emissions from ammonia production have decreased by 22 percent. Emissions in 2013 have increased by approximately 8 percent from the 2012 levels.

Table 4-18: CO_2 Emissions from Ammonia Production (MMT CO_2 Eq.)

Source	1990	2005	2009	2010	2011	2012	2013
Ammonia Production	13.0	9.2	8.5	9.2	9.3	9.4	10.2
Total	**13.0**	**9.2**	**8.5**	**9.2**	**9.3**	**9.4**	**10.2**

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values

Table 4-19: CO_2 Emissions from Ammonia Production (kt)

Source	1990	2005	2009	2010	2011	2012	2013
Ammonia Production	13,047	9,196	8,454	9,188	9,292	9,377	10,152
Total	**13,047**	**9,196**	**8,454**	**9,188**	**9,292**	**9,377**	**10,152**

Methodology

CO_2 emissions from production of synthetic ammonia from natural gas feedstock is based on the *2006 IPCC Guidelines* (IPCC 2006) Tier 1 and 2 method. A country-specific emission factor is developed and applied to national ammonia production to estimate emissions. The method uses a CO_2 emission factor published by the European Fertilizer Manufacturers Association (EFMA) that is based on natural gas-based ammonia production technologies that are similar to those employed in the United States. The CO_2 emission factor of 1.2 metric tons CO_2/metric ton NH_3 (EFMA 2000a) is applied to the percent of total annual domestic ammonia production from natural gas feedstock.

Emissions of CO_2 from ammonia production are then adjusted to account for the use of some of the CO_2 produced from ammonia production as a raw material in the production of urea. The CO_2 emissions reported for ammonia production are reduced by a factor of 0.733 multiplied by total annual domestic urea production. This corresponds to a stoichiometric CO_2/urea factor of 44/60, assuming complete conversion of NH_3 and CO_2 to urea (IPCC 2006, EFMA 2000b).

All synthetic ammonia production and subsequent urea production are assumed to be from the same process—conventional catalytic reforming of natural gas feedstock, with the exception of ammonia production from petroleum coke feedstock at one plant located in Kansas. Annual ammonia and urea production are shown in Table 4-20. The CO_2 emission factor for production of ammonia from petroleum coke is based on plant specific data, wherein all carbon contained in the petroleum coke feedstock that is not used for urea production is assumed to be emitted to the atmosphere as CO_2 (Bark 2004). Ammonia and urea are assumed to be manufactured in the same manufacturing complex, as both the raw materials needed for urea production are produced by the ammonia production process. The CO_2 emission factor of 3.57 metric tons CO_2/metric ton NH_3 for the petroleum coke feedstock process (Bark 2004) is applied to the percent of total annual domestic ammonia production from petroleum coke feedstock.

The emission factor of 1.2 metric ton CO_2/metric ton NH_3 for production of ammonia from natural gas feedstock was taken from the EFMA Best Available Techniques publication, Production of Ammonia (EFMA 2000a). The EFMA reported an emission factor range of 1.15 to 1.30 metric ton CO_2/metric ton NH_3, with 1.2 metric ton CO_2/metric ton NH_3 as a typical value (EFMA 2000a). Technologies (e.g., catalytic reforming process, etc.) associated with this factor are found to closely resemble those employed in the United States for use of natural gas as a feedstock. The EFMA reference also indicates that more than 99 percent of the CH_4 feedstock to the catalytic reforming process is ultimately converted to CO_2. The emission factor of 3.57 metric ton CO_2/metric ton NH_3 for production of ammonia from petroleum coke feedstock was developed from plant-specific ammonia production data and petroleum coke feedstock utilization data for the ammonia plant located in Kansas (Bark 2004). As noted earlier, emissions from fuels consumed for energy purposes during the production of ammonia are accounted for in the Energy chapter. The total ammonia production data for 2011, 2012, and 2013 were obtained from American Chemistry Council (2014). For years before 2011, ammonia production data (See Table 4-20) was obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, 2011, and 2012) and the Census Bureau of the U.S. Department of Commerce (U.S. Census Bureau 1991 through 1994, 1998 through 2010) as reported in Current Industrial Reports Fertilizer Materials and Related Products annual and quarterly reports. Urea-ammonia nitrate production from petroleum coke for years through 2011 was obtained from Coffeyville Resources (Coffeyville 2005, 2006, 2007a, 2007b, 2009, 2010, 2011, and 2012), and from CVR Energy, Inc. Annual Report (CVR 2012 and 2014) for 2012 and 2013. Urea production data for 1990 through 2008 were obtained from the Minerals Yearbook: Nitrogen (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Bureau of the Census (U.S. Bureau of the Census 2010 and 2011). The U.S. Bureau of the Census ceased collection of urea production statistics, and urea production data for 2011and 2012 were obtained from the Minerals Yearbook: Nitrogen (USGS 2014). The urea production data for 2013 are not yet published and so 2012 data has been used as proxy for 2013.

Table 4-20: Ammonia Production and Urea Production (kt)

Year	Ammonia Production	Urea Production
1990	15,425	7,450
2005	10,143	5,270
2009	9,372	5,084
2010	10,084	5,122
2011	10,325	5,430
2012	10,305	5,220
2013	10,930	5,220

Uncertainty and Time-Series Consistency

The uncertainties presented in this section are primarily due to how accurately the emission factor used represents an average across all ammonia plants using natural gas feedstock. Uncertainties are also associated with ammonia production estimates and the assumption that all ammonia production and subsequent urea production was from the same process—conventional catalytic reforming of natural gas feedstock, with the exception of one ammonia production plant located in Kansas that is manufacturing ammonia from petroleum coke feedstock. Uncertainty is also associated with the representativeness of the emission factor used for the petroleum coke-based ammonia process. It is also assumed that ammonia and urea are produced at collocated plants from the same natural gas raw material.

Recovery of CO_2 from ammonia production plants for purposes other than urea production (e.g., commercial sale, etc.) has not been considered in estimating the CO_2 emissions from ammonia production, as data concerning the disposition of recovered CO_2 are not available. Such recovery may or may not affect the overall estimate of CO_2 emissions depending upon the end use to which the recovered CO_2 is applied. Further research is required to determine whether byproduct CO_2 is being recovered from other ammonia production plants for application to end uses that are not accounted for elsewhere.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-21. Ammonia Production CO_2 emissions were estimated to be between 9.4 and 10.9 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 8 percent below and 8 percent above the emission estimate of 10.2 MMT CO_2 Eq.

Table 4-21: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Ammonia Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a]			
			(MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Ammonia Production	CO_2	10.2	9.4	10.9	-8%	+8%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Production estimates for urea production for the years 2011 and 2012 were updated using information obtained from the Minerals yearbook: Nitrogen (USGS 2014). This update resulted in an increase of emissions by approximately 3.5 percent in 2011 and 0.3 percent in 2012 emissions relative to the previous report.

Planned Improvements

Future improvements involve continuing to evaluate and analyze data reported under EPA's GHGRP to improve the emission estimates for the Ammonia Production source category. Particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[161] Specifically, the planned improvements include assessing data to update the emission factors to include both fuel and feedstock CO_2 emissions and incorporate CO_2 capture and storage. Methodologies will also be updated if additional ammonia-production plants are found to use hydrocarbons other than natural gas for ammonia production.

4.6 Urea Consumption for Non-Agricultural Purposes

Urea is produced using ammonia and CO_2 as raw materials. All urea produced in the United States is assumed to be produced at ammonia production facilities where both ammonia and CO_2 are generated. There are approximately 20 of these facilities operating in the United States.

The chemical reaction that produces urea is:

$$2NH_3 + CO_2 \rightarrow NH_2COONH_4 \rightarrow CO(NH_2)_2 + H_2O$$

This section accounts for CO_2 emissions associated with urea consumed exclusively for non-agricultural purposes. CO_2 emissions associated with urea consumed for fertilizer are accounted for in the Cropland Remaining Cropland section of the *Land Use, Land-Use Change, and Forestry* chapter.

Urea is used as a nitrogenous fertilizer for agricultural applications and also in a variety of industrial applications. Urea's industrial applications include its use in adhesives, binders, sealants, resins, fillers, analytical reagents, catalysts, intermediates, solvents, dyestuffs, fragrances, deodorizers, flavoring agents, humectants and dehydrating agents, formulation components, monomers, paint and coating additives, photosensitive agents, and surface treatments agents. In addition, urea is used for abating N_2O emissions from coal-fired power plants and diesel transportation motors.

Emissions of CO_2 from urea consumed for non-agricultural purposes in 2013 were estimated to be 4.7 MMT CO_2 Eq. (4,663 kt), and are summarized in Table 4-22 and Table 4-23. Net CO_2 emissions from urea consumption for non-agricultural purposes in 2013 have increased by approximately 23 percent from 1990.

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[161] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdffiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

Table 4-22: CO_2 Emissions from Urea Consumption for Non-Agricultural Purposes (MMT CO_2 Eq.)

Source	1990	2005	2009	2010	2011	2012	2013
Urea Consumption	3.8	3.7	3. 4	4.7	4.0	4.4	4.7
Total	**3.8**	**3.7**	**3.4**	**4.7**	**4.0**	**4.4**	**4.7**

Table 4-23: CO_2 Emissions from Urea Consumption for Non-Agricultural Purposes (kt)

Source	1990	2005	2009	2010	2011	2012	2013
Urea Consumption	3,784	3,653	3,427	4,730	4,029	4,449	4,663
Total	**3,784**	**3,653**	**3,427**	**4,730**	**4,029**	**4,449**	**4,663**

Methodology

Emissions of CO_2 resulting from urea consumption for non-agricultural purposes are estimated by multiplying the amount of urea consumed in the United States for non-agricultural purposes by a factor representing the amount of CO_2 used as a raw material to produce the urea. This method is based on the assumption that all of the carbon in urea is released into the environment as CO_2 during use, and consistent with the *2006 IPCC Guidelines* (IPCC 2006).

The amount of urea consumed for non-agricultural purposes in the United States is estimated by deducting the quantity of urea fertilizer applied to agricultural lands, which is obtained directly from the *Land Use, Land-Use Change, and Forestry* chapter (see Table 6-26) and is reported in Table 4-24, from the total domestic supply of urea. The domestic supply of urea is estimated based on the amount of urea produced plus the sum of net urea imports and exports. A factor of 0.73 tons of CO_2 per ton of urea consumed is then applied to the resulting supply of urea for non-agricultural purposes to estimate CO_2 emissions from the amount of urea consumed for non-agricultural purposes. The 0.733 tons of CO_2 per ton of urea emission factor is based on the stoichiometry of producing urea from ammonia and CO_2. This corresponds to a stoichiometric CO_2/urea factor of 44/60, assuming complete conversion of NH_3 and CO_2 to urea (IPCC 2006, EFMA 2000).

Urea production data for 1990 through 2008 were obtained from the Minerals Yearbook: Nitrogen (USGS 1994 through 2009). Urea production data for 2009 through 2010 were obtained from the U.S. Bureau of the Census (2011). The U.S. Bureau of the Census ceased collection of urea production statistics in 2011, therefore, urea production data for 2011and 2012 were obtained from the Minerals Yearbook: Nitrogen (USGS 2014). Urea production data for 2013 are not yet publicly available and so 2012 data has been used as proxy. Urea import data for 2011 and 2012 were taken from U.S. Fertilizer Import/Exports from USDA Economic Research Service Data Sets (U.S. Department of Agriculture 2012). Urea import data for the previous years were obtained from the U.S. Census Bureau Current Industrial Reports Fertilizer Materials and Related Products annual and quarterly reports for 1997 through 2010 U.S. Census Bureau (1998 through 2011), The Fertilizer Institute (TFI 2002) for 1993 through 1996, and the United States International Trade Commission Interactive Tariff and Trade DataWeb (U.S. ITC 2002) for 1990 through 1992 (see Table 4-24). Urea export data for 1990 through 2012 were taken from U.S. Fertilizer Import/Exports from USDA Economic Research Service Data Sets (U.S. Department of Agriculture 2012). Urea exports and imports data for 2013 is not yet available and so 2012 data has been used as proxy.

Table 4-24: Urea Production, Urea Applied as Fertilizer, Urea Imports, and Urea Exports (kt)

Year	Urea Production	Urea Applied as Fertilizer	Urea Imports	Urea Exports
1990	7,450	3,296	1,860	854
2005	5,270	4,779	5,026	536
2009	5,084	4,848	4,727	289
2010	5,122	5,152	6,631	152
2011	5,430	5,589	5,860	207
2012	5,220	5,762	6,944	336
2013	5,220	5,469	6,944	336

Uncertainty and Time-Series Consistency

There is limited publicly-available data on the quantities of urea produced and consumed for non-agricultural purposes. Therefore, the amount of urea used for non-agricultural purposes is estimated based on a balance that relies on estimates of urea production, urea imports, urea exports, and the amount of urea used as fertilizer. The primary uncertainties associated with this source category are associated with the accuracy of these estimates as well as the fact that each estimate is obtained from a different data source. Because urea production estimates are no longer available from the USGS, there is additional uncertainty associated with urea produced beginning in 2011. There is also uncertainty associated with the assumption that all of the carbon in urea is released into the environment as CO_2 during use.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-25. CO_2 emissions associated with urea consumption for non-agricultural purposes were estimated to be between 4.2 and 5.1 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 10 percent below and 10 percent above the emission estimate of 4.7 MMT CO_2 Eq.

Table 4-25: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Urea Consumption for Non-Agricultural Purposes (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Urea Consumption for Non-Agricultural Purposes	CO_2	4.7	4.2	5.1	-10%	+10%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Production estimates for urea production for the years 2011 and 2012 were updated using information obtained from the Minerals Yearbook: Nitrogen (USGS 2014). Also, the amount of urea consumed for agricultural purposes in the United States for 2012 was revised based on the most recent data obtained from the *Land Use, Land-Use Change, and Forestry* chapter (see Table 6-26). These updates resulted in an increase of emissions by approximately 1 percent in 2011 and a decrease of approximately 15 percent in 2012 emissions.

4.7 Nitric Acid Production (IPCC Source Category 2B2)

Nitrous oxide (N_2O) is emitted during the production of nitric acid (HNO_3), an inorganic compound used primarily to make synthetic commercial fertilizers. It is also a major component in the production of adipic acid—a feedstock for nylon—and explosives. Virtually all of the nitric acid produced in the United States is manufactured by the catalytic oxidation of ammonia (EPA 1997). There are two different nitric acid production methods: weak nitric acid and high-strength nitric acid. The first method utilizes oxidation, condensation, and absorption to produce nitric acid at concentrations between 30 and 70 percent nitric acid. High-strength acid (90 percent or greater nitric acid) can be produced from dehydrating, bleaching, condensing, and absorption of the weak nitric acid. The basic process technology for producing nitric acid has not changed significantly over time. Most U.S. plants were built between 1960 and 2000. As of 2013, there are 35 active weak nitric acid production plants and one high-strength nitric acid production plant in U.S. (EPA 2010b; EPA 2014).

During this reaction, N_2O is formed as a byproduct and is released from reactor vents into the atmosphere. Emissions from fuels consumed for energy purposes during the production of nitric acid are accounted for in the Energy chapter.

Nitric acid is made from the reaction of ammonia (NH_3) with oxygen (O_2) in two stages. The overall reaction is:

$$4NH_3 + 8O_2 \rightarrow 4HNO_3 + 4H_2O$$

Currently, the nitric acid industry controls emissions of NO and NO_2 (i.e., NO_x). As such, the industry in the United States uses a combination of non-selective catalytic reduction (NSCR) and selective catalytic reduction (SCR) technologies. In the process of destroying NO_x, NSCR systems are also very effective at destroying N_2O. However, NSCR units are generally not preferred in modern plants because of high energy costs and associated high gas temperatures. NSCR systems were installed in nitric plants built between 1971 and 1977, approximately one-third of the weak acid production plants have NSCRs. U.S. facilities are using both tertiary (e.g., NSCR) and secondary controls (alternate catalysts).

N_2O emissions from this source were estimated to be 10.7 MMT CO_2 Eq. (36 kt of N_2O) in 2013 (see Table 4-26). Emissions from nitric acid production have decreased by 12 percent since 1990, with the trend in the time series closely tracking the changes in production. Emissions have decreased by 26 percent since 1997, the highest year of production in the time series.

Table 4-26: N_2O Emissions from Nitric Acid Production (MMT CO_2 Eq. and kt N_2O)

Year	MMT CO_2 Eq.	kt N_2O
1990	12.1	41
2005	11.3	38
2009	9.6	32
2010	11.5	39
2011	10.9	37
2012	10.5	35
2013	10.7	36

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

Methodology

Emissions of N_2O were calculated using the estimation methods provided by the *2006 IPCC Guidelines* (IPCC 2006) and country specific methods from N_2O EPA's Greenhouse Gas Reporting Program. The *2006 IPCC*

Guidelines Tier 2 method was used to estimate emissions from nitric acid production for 1990 through 2009, and a country specific approach similar to the IPCC Tier 3 method was used to estimate N_2O emissions for 2010 through 2013.

2010 through 2013

Process N_2O emissions and nitric acid production data were obtained directly from EPA's GHGRP for 2010 through 2013 by aggregating reported facility-level data (EPA 2014). In the United States, all nitric acid facilities producing weak nitric acid (30-70 percent in strength) are required to report annual GHG emissions data to EPA as per the requirements of its Greenhouse Gas Reporting Program (GHGRP). As of 2013, there are 35 facilities that report to EPA, including the known single high-strength nitric acid production facility in the United States (EPA 2014). All nitric acid (weak acid) facilities are required to calculate process emissions using a site-specific emission factor developed through annual performance testing under typical operating conditions or by directly measuring N_2O emissions using monitoring equipment. The high-strength nitric acid facility also reports N_2O emissions associated with weak acid production and this may capture all relevant emissions, pending additional further EPA research. More details on the calculation and monitoring methods applicable to Nitric Acid facilities can be found under Subpart V: Nitric Acid Production of the regulation, Part 98.[162]

1990 through 2009

Using the GHGRP data for 2010,[163] country-specific N_2O emission factors were calculated for nitric acid production with abatement and without abatement (i.e., controlled and uncontrolled emission factors). These emission factors were used to estimate N_2O emissions from nitric acid production for years prior to the GHGRP data (i.e., 1990 through 2009): 3.3 kg N_2O/metric ton HNO_3 produced at plants using abatement technologies (e.g., tertiary systems such as NSCR systems) and 5.98 kg N_2O/metric ton HNO_3 produced at plants not equipped with abatement technology. Based on the available data, it was assumed that emission factors for 2010 would be more representative of abatement application in 1990 through 2009. Initial review of historical data indicates that percent production with and without abatement change over time and also year over year due to changes in application of facility-level abatement technologies, maintenance of abatement technologies, and also due to plant closures and start-ups (EPA 2010a, 2012, 2013b; Desai 2012; CAR 2013). The installation dates of N_2O abatement technologies are not known at most facilities, but it is assumed that facilities reporting abatement technology use have had this technology installed and operational for the duration of the time series considered in this report (especially NSCRs).

The country-specific N_2O emission factors were used in conjunction with annual production and national share of production with and without abatement technologies to estimate N_2O emissions for 1990 through 2009, using the following equation:

$$E_i = \left[\left(P_i \times \%P_{C,i} \times EF_c\right) + \left(P_i \times \%P_{unc,i} \times EF_{unc}\right)\right]$$

where,

E_i = Annual N_2O Emissions for year i (kg/yr)
P_i = Annual nitric acid production for year i (metric tons HNO_3)
$\%P_{c,i}$ = Percent national production of HNO_3 with N_2O abatement technology (%)
EF_c = N_2O emission factor, with abatement technology (kg N_2O/metric ton HNO_3)
$\%P_{unc,i}$ = Percent national production of HNO_3 without N_2O abatement technology (%)
EF_{unc} = N_2O emission factor, without abatement technology (kg N_2O/metric ton HNO_3)
i = year from 1990 through 2009

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[162] Located at <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl>.

[163] National N_2O process emissions, national production, and national share of nitric acid production with abatement and without abatement technology was aggregated from the GHGRP facility-level data for 2010-2013 (i.e., percent production with and without abatement).

Nitric acid production data for the United States for 1990 through 2009 were obtained from the U.S. Census Bureau (U.S. Census Bureau 2008 through 2010) (see Table 4-27). Publicly-available information on plant-level abatement technologies was used to estimate the shares of nitric acid production with and without abatement for 2008 and 2009 (EPA 2010a, 2012, 2013b; Desai 2012; CAR 2013). Publicly-available data on use of abatement technologies were not available for 1990-2007. Therefore, the share of national production with and without abatement for 2008 was assumed to be constant for 1990 through 2007.

Table 4-27: Nitric Acid Production (kt)

Year	kt
1990	7,195
2005	6,711
2009	5,924
2010	7,444
2011	7,606
2012	7,453
2013	7,572

Uncertainty and Time-Series Consistency

Uncertainty associated with the parameters used to estimate N_2O emissions includes that of production data, the share of U.S. nitric acid production attributable to each emission abatement technology over the time series (especially prior to 2010), and the associated emission factors applied to each abatement technology type. While some information has been obtained through outreach with industry associations, limited information is available over the time series (especially prior to 2010) for a variety of facility level variables, including plant specific production levels, plant production technology (e.g., low, high pressure, etc.), and abatement technology type, installation date of abatement technology, and accurate destruction and removal efficiency rates.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-28. N_2O emissions from nitric acid production were estimated were estimated to be between 10.1 and 11.3 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below to 5 percent above the 2013 emissions estimate of 10.7 MMT CO_2 Eq.

Table 4-28: Approach 2 Quantitative Uncertainty Estimates for N_2O Emissions from Nitric Acid Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a]			
			(MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Nitric Acid Production	N_2O	10.7	10.1	11.3	-5%	+5%

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2-equivalent emissions from CH_4, HFCs, and PFCs. The GWPs of N_2O and SF_6 have decreased, leading to a decrease in CO_2-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

In addition, GHGRP data from subpart V of regulation 40 CFR Part 98 were used to recalculate emissions from nitric acid production over the entire time series (EPA 2014), and used directly for emission estimates for 2010 through 2013. Nitric acid production and N_2O emissions data were available for 2010 through 2013 from EPA's GHGRP, given nearly all nitric acid production facilities, with the exception of the strong acid facility, in the United States are required to report annual data under subpart V. Country-specific N_2O emission factors were developed using the 2010 GHGRP emissions and production data for nitric acid production with abatement and without abatement. Due to differences in operational efficiencies and recent installation of abatement technology at some U.S. facilities, 2010 GHGRP production data were used for recalculating time series emissions (1990 through 2009) instead of average factors developed from 2010 through 2013 GHGRP data. As per the 2010 GHGRP data, 70.7 percent of total domestic nitric acid production was estimated to be produced without any abatement.

Using the 2010 GHGRP data, emission factors for production with abatement and without abatement were calculated to be 3.3 kg N_2O/metric ton nitric acid produced and 5.98 kg N_2O/metric ton nitric acid produced, respectively. These emission factors and historical production data from the U.S. Census Bureau were used to calculate emissions for 1990 through 2009. The emission factors were used in conjunction with existing estimates on the share of production with and without N_2O abatement technology to estimate N_2O emissions for 1990 through 2009.

For 2009, an estimated 19.7 percent of nitric acid production was produced using N_2O abatement technology and 80.3 percent production was without abatement technology (EPA 2010a, 2013b, 2012; Desai 2012; CAR 2013). Similarly for 2008, an estimated 12.3 percent of nitric acid production was without abatement and 87.7 percent production was with abatement technology (EPA 2012). Since data on the use of abatement technology was not publicly available for 1990 through 2007, the national shares of production with and without abatement for 2008 were used for all prior years (i.e., 1990 through 2007).

Time series emissions for 1990 through 2009 were recalculated, and the revised emission estimates are approximately 30 percent lower than the prior estimates.

4.8 Adipic Acid Production (IPCC Source Category 2B3)

Adipic acid is produced through a two-stage process during which N_2O is generated in the second stage. Emissions from fuels consumed for energy purposes during the production of adipic acid are accounted for in the Energy chapter. The first stage of manufacturing usually involves the oxidation of cyclohexane to form a cyclohexanone/ cyclohexanol mixture. The second stage involves oxidizing this mixture with nitric acid to produce adipic acid. N_2O is generated as a byproduct of the nitric acid oxidation stage and is emitted in the waste gas stream (Thiemens and Trogler 1991). The second stage is represented by the following chemical reaction:

$$(CH_2)_5CO(cyclohexanone) + (CH_2)_5CHOH(cyclohexanol) + wHNO_3 \\ \rightarrow HOOC(CH_2)_4COOH(adipic\ acid) + xN_2O + yH_2O$$

Process emissions from the production of adipic acid vary with the types of technologies and level of emission controls employed by a facility. In 1990, two major adipic acid-producing plants had N_2O abatement technologies in place and, as of 1998, three major adipic acid production facilities had control systems in place (Reimer et al. 1999). One small plant, which last operated in April 2006 and represented approximately two percent of production, did not control for N_2O (VA DEQ 2009; ICIS 2007; VA DEQ 2006). In 2013, catalytic reduction, non-selective catalytic reduction (NSCR) and thermal reduction abatement technologies were applied as N_2O abatement measures at adipic acid facilities (EPA 2014).

Worldwide, only a few adipic acid plants exist. The United States, Europe, and China are the major producers. In 2013, the United States had two companies with a total of three adipic acid production facilities (two in Texas and one in Florida), all of which were operational (EPA 2014). The United States accounts for the largest share of global adipic acid production capacity (30 percent), followed by the European Union (29 percent) and China (22 percent) (SEI 2010). Adipic acid is a white crystalline solid used in the manufacture of synthetic fibers, plastics, coatings, urethane foams, elastomers, and synthetic lubricants. Commercially, it is the most important of the aliphatic dicarboxylic acids, which are used to manufacture polyesters. Eighty-four percent of all adipic acid produced in the United States is used in the production of nylon 6,6; 9 percent is used in the production of polyester polyols; 4 percent is used in the production of plasticizers; and the remaining 4 percent is accounted for by other uses, including unsaturated polyester resins and food applications (ICIS 2007). Food grade adipic acid is used to provide some foods with a "tangy" flavor (Thiemens and Trogler 1991).

N_2O emissions from adipic acid production were estimated to be 4.0 MMT CO_2 Eq. (13 kt) in 2013 (see Table 4-29). National adipic acid production has increased by approximately 11 percent over the period of 1990 through 2013, to approximately 840,000 metric tons (ACC 2014). Over the period 1990 to 2013, emissions have been reduced by 74 percent due to both the widespread installation of pollution control measures in the late 1990s and plant idling in the late 2000s. In April 2006, the smallest of the four facilities ceased production of adipic acid (VA DEQ 2009); furthermore, one of the major adipic acid production facilities was not operational in 2009 or 2010 (Desai 2010). All three remaining facilities were in operation in 2013. Very little information on annual trends in the activity data exist for adipic acid.

Table 4-29: N_2O Emissions from Adipic Acid Production (MMT CO_2 Eq. and kt N_2O)

Year	MMT CO_2 Eq.	kt N_2O
1990	15.2	51
2005	7.1	24
2009	2.7	9
2010	4.2	14
2011	10.2	34
2012	5.5	19
2013	4.0	13

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

Methodology

Emissions are estimated using both Tier 2 and Tier 3 methods consistent with consistent the *2006 IPCC Guidelines* (IPCC 2006). Facility level greenhouse gas emissions data were obtained from the GHGRP for the years 2010 through 2013 (EPA 2014) and aggregated to national N_2O emissions. Consistent with IPCC Tier 3 methods, all adipic acid production facilities are required to calculate emissions using a facility-specific emission factor developed through annual performance testing under typical operating conditions or by directly measuring N_2O emissions using monitoring equipment. More information on the monitoring methods for process N_2O emissions applicable to adipic acid production facilities under Subpart E can be found in the electronic code of federal regulations.[164]

Due to confidential business information, plant names are not provided in this section. Therefore, the four adipic acid-producing facilities will be referred to as Plants 1 through 4. Plant 4 was closed in April 2006. Overall, as noted

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[164] See <http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl>.

above, the three plants that are currently operating facilities use abatement equipment. Plants 1 and 2 employ catalytic destruction and Plant 3 employs thermal destruction.

2010 through 2013

All emission estimates for 2010 through 2013 were obtained through analysis of the GHGRP data (EPA 2014), which is consistent with the *2006 IPCC Guidelines* (IPCC 2006) Tier 3 method.

1990 through 2009

For years prior to EPA's GHGRP reporting, for both Plants 1 and 2, 1990 to 2009 emission estimates were obtained directly from the plant engineer and account for reductions due to control systems in place at these plants during the time series. These prior estimates are considered confidential business information and hence are not published (Desai 2010). These estimates were based on continuous process monitoring equipment installed at the two facilities. In 2009 and 2010, no adipic acid production occurred at Plant 1 per reporting to EPA's GHGRP (EPA 2012; Desai 2011b).

For the Plant 4, 1990 through 2009 N_2O emissions were estimated using the following Tier 2 equation from the *2006 IPCC Guidelines* until shutdown of the plant in 2006:

$$E_{aa} = Q_{aa} \times EF_{aa} \times (1 - [DF \times UF])$$

where,

E_{aa} = N_2O emissions from adipic acid production, metric tons
Q_{aa} = Quantity of adipic acid produced, metric tons
EF_{aa} = Emission factor, metric ton N_2O/metric ton adipic acid produced
DF = N_2O destruction factor
UF = Abatement system utility factor

The adipic acid production is multiplied by an emission factor (i.e., N_2O emitted per unit of adipic acid produced), which has been estimated, based on experiments that the reaction stoichiometry for N_2O production in the preparation of adipic acid at approximately 0.3 metric tons of N_2O per metric ton of product (IPCC 2006). The "N_2O destruction factor" in the equation represents the percentage of N_2O emissions that are destroyed by the installed abatement technology. The "abatement system utility factor" represents the percentage of time that the abatement equipment operates during the annual production period. No abatement equipment was installed the Inolex/Allied Signal facility, which last operated in April 2006 (VA DEQ 2009). Plant-specific production data for this facility were obtained across the time series from 1990 through 2006 from the Virginia Department of Environmental Quality (VA DEQ 2010). The plant-specific production data were then used for calculating emissions as described above.

For Plant 3, 2005 through 2009 emissions were obtained directly from the plant (Desai 2011a). For 1990 through 2004, emissions were estimated using plant-specific production data and IPCC factors as described above for Plant 4. Plant-level adipic acid production for 1990 through 2003 was estimated by allocating national adipic acid production data to the plant level using the ratio of known plant capacity to total national capacity for all U.S. plants (ACC 2014; CMR 2001, 1998; CW 1999; C&EN 1995, 1994, 1993, and 1992). For 2004, actual plant production data were obtained and used for emission calculations (CW 2005).

Plant capacities for 1990 through 1994 were obtained from Chemical and Engineering News, "Facts and Figures" and "Production of Top 50 Chemicals" (C&EN 1992 through 1995). Plant capacities for 1995 and 1996 were kept the same as 1994 data. The 1997 plant capacities were taken from Chemical Market Reporter "Chemical Profile: Adipic Acid" (CMR 1998). The 1998 plant capacities for all four plants and 1999 plant capacities for three of the plants were obtained from Chemical Week, Product Focus: Adipic Acid/Adiponitrile (CW 1999). Plant capacities for 2000 for three of the plants were updated using Chemical Market Reporter, "Chemical Profile: Adipic Acid" (CMR 2001). For 2001 through 2003, the plant capacities for three plants were kept the same as the year 2000 capacities. Plant capacity for 1999 to 2003 for the one remaining plant was kept the same as 1998.

National adipic acid production data (see Table 4-30) from 1990 through 2013 were obtained from the American Chemistry Council (ACC 2014).

Table 4-30: Adipic Acid Production (kt)

Year	kt
1990	755
2005	865
2009	650
2010	720
2011	810
2012	810
2013	840

Uncertainty and Time-Series Consistency

Uncertainty associated with N_2O emission estimates includes the methods used by companies to monitor and estimate emissions. While some information has been obtained through outreach with facilities, limited information is available over the time series on these methods, but also abatement technology destruction and removal efficiency rates and plant specific production levels.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-31. N_2O emissions from adipic acid production for 2013 were estimated to be between 3.8 and 4.2 MMT CO_2 Eq. at the 95 percent confidence level. These values indicate a range of approximately 4 percent below to 4 percent above the 2013 emission estimate of 4.0 MMT CO_2 Eq.

Table 4-31: Approach 2 Quantitative Uncertainty Estimates for N_2O Emissions from Adipic Acid Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Adipic Acid Production	N_2O	4.0	3.8	4.2	-4%	+4%

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2-equivalent emissions from CH_4, HFCs, and PFCs. The GWPs of N_2O and SF_6 have decreased, leading to a decrease in CO_2-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

4.9 Silicon Carbide Production and Consumption (IPCC Source Category 2B5)

Carbon dioxide (CO_2) and methane (CH_4) are emitted from the production of silicon carbide (SiC), a material used as an industrial abrasive. Silicon carbide is produced for abrasive, metallurgical, and other non-abrasive applications in the United States. Production for metallurgical and other non-abrasive applications is not available and therefore both CO_2 and CH_4 estimates are based solely upon production estimates of silicon carbide for abrasive applications. Emissions from fuels consumed for energy purposes during the production of silicon carbide are accounted for in the Energy chapter.

To produce SiC, silica sand or quartz (SiO_2) is reacted with carbon in the form of petroleum coke. A portion (about 35 percent) of the carbon contained in the petroleum coke is retained in the SiC. The remaining carbon is emitted as CO_2, CH_4, or CO. The overall reaction is shown below (but in practice it does not proceed according to stoichiometry):

$$SiO_2 + 3C \rightarrow SiC + 2CO \ (+ O_2 \rightarrow 2CO_2)$$

Carbon dioxide is also emitted from the consumption of SiC for metallurgical and other non-abrasive applications.

Markets for manufactured abrasives, including SiC, are heavily influenced by activity in the U.S. manufacturing sector, especially in the aerospace, automotive, furniture, housing, and steel manufacturing sectors. The USGS reports that a portion (approximately 50 percent) of SiC is used in metallurgical and other non-abrasive applications, primarily in iron and steel production (USGS 2006a). As a result of the economic downturn in 2008 and 2009, demand for SiC decreased in those years. Low cost imports, particularly from China, combined with high relative operating costs for domestic producers, continue to put downward pressure on the production of SiC in the United States. However, demand for SiC consumption in the United States has recovered somewhat from its lows in 2009 (USGS 2012a). Silicon carbide is manufactured at a single facility located in Illinois (USGS 2013b).

Carbon dioxide emissions from SiC production and consumption in 2013 were 0.17 MMT CO_2 Eq. (169 kt). Approximately 54 percent of these emissions resulted from SiC production while the remainder resulted from SiC consumption. Methane emissions from SiC production in 2013 were 0.01 MMT CO_2 Eq. (0.4 kt CH_4) (see Table 4-32: and Table 4-33). Emissions have fluctuated in recent years, but 2013 emissions are only about 45 percent of emissions in 1990.

Table 4-32: CO_2 and CH_4 Emissions from Silicon Carbide Production and Consumption (MMT CO_2 Eq.)

Year	**1990**	**2005**	**2009**	**2010**	**2011**	**2012**	**2013**
CO_2	0.4	0.2	0.1	0.2	0.2	0.2	0.2
CH_4	+	+	+	+	+	+	+
Total	**0.4**	**0.2**	**0.1**	**0.2**	**0.2**	**0.2**	**0.2**

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-33: CO_2 and CH_4 Emissions from Silicon Carbide Production and Consumption (kt)

Year	**1990**	**2005**	**2009**	**2010**	**2011**	**2012**	**2013**
CO_2	375	219	145	181	170	158	169
CH_4	1	+	+	+	+	+	+

+ Does not exceed 0.5 kt.

Methodology

Emissions of CO_2 and CH_4 from the production of SiC were calculated using the Tier 1 method provided by the *2006 IPCC Guidelines* (IPCC 2006). Annual estimates of SiC production were multiplied by the appropriate emission factor, as shown below:

$$E_{sc,CO2} = EF_{sc,CO2} \times Q_{sc}$$

$$E_{sc,CH4} = EF_{sc,CH4} \times Q_{sc} \times \left(\frac{1\ metric\ ton}{1000\ kg}\right)$$

where,

$E_{sc,CO2}$ = CO_2 emissions from production of SiC, metric tons
$E_{sc,CO2}$ = Emission factor for production of SiC, metric ton CO_2/metric ton SiC
Q_{sc} = Quantity of SiC produced, metric tons
$E_{sc,CH4}$ = CH_4 emissions from production of SiC, metric tons
$E_{sc,CH4}$ = Emission factor for production of SiC, kilogram CH_4/metric ton SiC

Emission factors were taken from the *2006 IPCC Guidelines* (IPCC 2006):

- 2.62 metric tons CO_2/metric ton SiC
- 11.6 kg CH_4/metric ton SiC

Emissions of CO_2 from silicon carbide consumption for metallurgical uses were calculated by multiplying the annual utilization of SiC for metallurgical uses (reported annually in the USGS Minerals Yearbook for Silicon) by the carbon content of SiC (31.5 percent), which was determined according to the molecular weight ratio of SiC.

Emissions of CO_2 from silicon carbide consumption for other non-abrasive uses were calculated by multiplying the annual SiC consumption for non-abrasive uses by the carbon content of SiC (31.5 percent). The annual SiC consumption for non-abrasive uses was calculated by multiplying the annual SiC consumption (production plus net imports) by the percent used in metallurgical and other non-abrasive uses (50 percent) (USGS 2006a) and then subtracting the SiC consumption for metallurgical use.

Production data for 1990 through 2012 were obtained from the Minerals Yearbook: Manufactured Abrasives (USGS 1991a through 2013a). Production data for 2013 were obtained from the Minerals Industry Surveys: Abrasives (Manufactured) (USGS 2014). Silicon carbide consumption by major end use was obtained from the Minerals Yearbook: Silicon (USGS 1991b through 2011b, 2012c, and 2013b) (see Table 4-34). Net imports for the entire time series were obtained from the U.S. Census Bureau (2005 through 2014).

Table 4-34: Production and Consumption of Silicon Carbide (Metric Tons)

Year	Production	Consumption
1990	105,000	172,465
2005	35,000	220,149
2009	35,000	92,280
2010	35,000	154,540
2011	35,000	136,222
2012	35,000	114,265
2013	35,000	134,054

Uncertainty and Time-Series Consistency

There is uncertainty associated with the emission factors used because they are based on stoichiometry as opposed to monitoring of actual SiC production plants. An alternative would be to calculate emissions based on the quantity of petroleum coke used during the production process rather than on the amount of silicon carbide produced. However, these data were not available. For CH_4, there is also uncertainty associated with the hydrogen-containing volatile compounds in the petroleum coke (IPCC 2006). There is also uncertainty associated with the use or destruction of methane generated from the process in addition to uncertainty associated with levels of production, net imports, consumption levels, and the percent of total consumption that is attributed to metallurgical and other non-abrasive uses.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-35. Silicon carbide production and consumption CO_2 emissions were estimated to be between 9 percent below and 9 percent above the emission estimate of 0.17 MMT CO_2 Eq. at the 95 percent confidence level. Silicon carbide production CH_4 emissions were estimated to be between 9 percent below and 10 percent above the emission estimate of 0.01 MMT CO_2 Eq. at the 95 percent confidence level.

Table 4-35: Approach 2 Quantitative Uncertainty Estimates for CH_4 and CO_2 Emissions from Silicon Carbide Production and Consumption (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a]			
			(MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Silicon Carbide Production and Consumption	CO_2	0.17	0.15	0.18	-9%	+9%
Silicon Carbide Production	CH_4	+	+	+	-9%	+10%

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

+ Does not exceed 0.05 MMT CO_2 Eq.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2-equivalent emissions from CH_4, HFCs, and PFCs. The GWPs of N_2O and SF_6 have decreased, leading to a decrease in CO_2-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter. This change caused a slight increase of emissions over the entire time series relative to the previous report.

Planned Improvements

Future improvements involve continuing to evaluate and analyze data reported under EPA's GHGRP to improve the emission estimates for the Silicon Carbide Production source category. Particular attention will be made to ensure time series consistency of the emission estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[165] In addition, improvements will involve continued research to determine if calcium carbide production and consumption data are available for the United States. If these data are available, calcium carbide emission estimates will be included in this source category.

4.10 Titanium Dioxide Production (IPCC Source Category 2B6)

Titanium dioxide (TiO_2) is manufactured using one of two processes: the chloride process and the sulfate process. The chloride process uses petroleum coke and chlorine as raw materials and emits process-related CO_2. Emissions from fuels consumed for energy purposes during the production of titanium dioxide are accounted for in the Energy chapter. The chloride process is based on the following chemical reactions:

$$2FeTiO_3 + 7Cl_2 + 3C \rightarrow 2TiCl_4 + 2FeCl_3 + 3CO_2$$

$$2TiCl_4 + 2O_2 \rightarrow 2TiO_2 + 4Cl_2$$

The sulfate process does not use petroleum coke or other forms of carbon as a raw material and does not emit CO_2.

The carbon in the first chemical reaction is provided by petroleum coke, which is oxidized in the presence of the chlorine and $FeTiO_3$ (rutile ore) to form CO_2. Since 2004, all TiO_2 produced in the United States has been produced using the chloride process, and a special grade of "calcined" petroleum coke is manufactured specifically for this purpose.

The principal use of TiO_2 is as a pigment in white paint, lacquers, and varnishes; it is also used as a pigment in the manufacture of paper, foods, plastics, and other products. In 2013, U.S. TiO_2 production totaled 1,200,000 metric tons (USGS 2014b). There were a total 6 plants producing TiO_2 in the United States—2 located in Mississippi, and single plants located in Delaware, Louisiana, Ohio, and Tennessee.

Emissions of CO_2 in 2013 were 1.6 MMT CO_2 Eq. (1,608 kt), which represents an increase of 35 percent since 1990 (see Table 4-36).

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[165] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

Table 4-36: CO_2 Emissions from Titanium Dioxide (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.2	1,195
2005	1.8	1,755
2009	1.6	1,648
2010	1.8	1,769
2011	1.7	1,729
2012	1.5	1,528
2013	1.6	1,608

Methodology

Emissions of CO_2 from TiO_2 production were calculated by multiplying annual national TiO_2 production by chloride-process-specific emission factors using a Tier 1 approach provided in *2006 IPCC Guidelines* (IPCC 2006). The Tier 1 equation is as follows:

$$E_{td} = EF_{td} \times Q_{td}$$

where,

E_{td} = CO_2 emissions from TiO_2 production, metric tons
EF_{td} = Emission factor (chloride process), metric ton CO_2/metric ton TiO_2
Q_{td} = Quantity of TiO_2 produced

Data were obtained for the total amount of TiO_2 produced each year. For years prior to 2004, it was assumed that TiO_2 was produced using the chloride process and the sulfate process in the same ratio as the ratio of the total U.S. production capacity for each process. As of 2004, the last remaining sulfate-process plant in the United States closed; therefore, 100 percent of post-2004 production uses the chloride process (USGS 2005). The percentage of production from the chloride process is estimated at 100 percent since 2004. An emission factor of 1.34 metric tons CO_2/metric ton TiO_2 was applied to the estimated chloride-process production (IPCC 2006). It was assumed that all TiO_2 produced using the chloride process was produced using petroleum coke, although some TiO_2 may have been produced with graphite or other carbon inputs.

The emission factor for the TiO_2 chloride process was taken from the *2006 IPCC Guidelines* (IPCC 2006). Titanium dioxide production data and the percentage of total TiO_2 production capacity that is chloride process for 1990 through 2012 (see Table 4-37:) were obtained through the Minerals Yearbook: Titanium Annual Report (USGS 1991 through 2014a). Production data for 2013 was obtained from the Minerals Commodity Summary: Titanium and Titanium Dioxide (USGS 2014b). Data on the percentage of total TiO_2 production capacity that is chloride process were not available for 1990 through 1993, so data from the 1994 USGS Minerals Yearbook were used for these years. Because a sulfate process plant closed in September 2001, the chloride process percentage for 2001 was estimated based on a discussion with Joseph Gambogi (2002). By 2002, only one sulfate plant remained online in the United States and this plant closed in 2004 (USGS 2005).

Table 4-37: Titanium Dioxide Production (kt)

Year	kt
1990	979
2005	1,310
2009	1,230
2010	1,320
2011	1,290
2012	1,140

2013	1,200

Uncertainty and Time-Series Consistency

Each year, USGS collects titanium industry data for titanium mineral and pigment production operations. If TiO_2 pigment plants do not respond, production from the operations is estimated on the basis of prior year production levels and industry trends. Variability in response rates varies from 67 to 100 percent of TiO_2 pigment plants over the time series.

Although some TiO_2 may be produced using graphite or other carbon inputs, information and data regarding these practices were not available. Titanium dioxide produced using graphite inputs, for example, may generate differing amounts of CO_2 per unit of TiO_2 produced as compared to that generated through the use of petroleum coke in production. While the most accurate method to estimate emissions would be to base calculations on the amount of reducing agent used in each process rather than on the amount of TiO_2 produced, sufficient data were not available to do so.

As of 2004, the last remaining sulfate-process plant in the United States closed. Since annual TiO_2 production was not reported by USGS by the type of production process used (chloride or sulfate) prior to 2004 and only the percentage of total production capacity by process was reported, the percent of total TiO_2 production capacity that was attributed to the chloride process was multiplied by total TiO_2 production to estimate the amount of TiO_2 produced using the chloride process. Finally, the emission factor was applied uniformly to all chloride-process production, and no data were available to account for differences in production efficiency among chloride-process plants. In calculating the amount of petroleum coke consumed in chloride-process TiO_2 production, literature data were used for petroleum coke composition. Certain grades of petroleum coke are manufactured specifically for use in the TiO_2 chloride process; however, this composition information was not available.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-38: Titanium dioxide consumption CO_2 emissions were estimated to be between 1.4 and 1.8 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 13 percent below and 13 percent above the emission estimate of 1.6 MMT CO_2 Eq.

Table 4-38: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Titanium Dioxide Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a]			
			(MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Titanium Dioxide Production	CO_2	1.6	1.4	1.8	-13%	+13%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Production data for 2012 were updated relative to the previous Inventory based on recently published data in the USGS Minerals Yearbook: Titanium 2012 (USGS 2014a). This resulted in a 12 percent decrease in 2012 CO_2 emissions from TiO_2 production relative to the previous report.

Planned Improvements

Pending resources, a potential improvement to the Inventory estimates for this source category would include the derivation of country-specific emission factors, based on annual data reported under EPA's GHGRP for 2010 through 2013 (i.e. aggregated emissions and titanium production). Information on titanium dioxide production is collected by EPA's GHGRP for all facilities for years 2010 through 2013 and would also have to be assessed against criteria EPA has established to publish aggregated confidential business information (CBI) reported under EPA's GHGRP. In order to provide estimates for the entire time series (i.e., 1990 through 2009), the applicability of more recent GHGRP data to previous years' estimates will need to be evaluated, and additional data that could be utilized in the calculations for this source category may need to be researched. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[166]

In addition, the planned improvements include researching the significance of titanium-slag production in electric furnaces and synthetic-rutile production using the Becher process in the United States. Significant use of these production processes will be included in future estimates.

4.11 Soda Ash Production and Consumption (IPCC Source Category 2B7)

Carbon dioxide is generated as a byproduct of calcining trona ore to produce soda ash, and is eventually emitted into the atmosphere. In addition, CO_2 may also be released when soda ash is consumed. Emissions from fuels consumed for energy purposes during the production and consumption of soda ash are accounted for in the Energy sector.

Calcining involves placing crushed trona ore into a kiln to convert sodium bicarbonate into crude sodium carbonate that will later be filtered into pure soda ash. The emission of CO_2 during trona-based production is based on the following reaction:

$$2Na2CO_3 \bullet NaHCO_3 \bullet 2H_2O(Trona) \rightarrow 3Na2CO_3(Soda\ Ash) + 5H_2O + CO_2$$

Soda ash (sodium carbonate, Na_2CO_3) is a white crystalline solid that is readily soluble in water and strongly alkaline. Commercial soda ash is used as a raw material in a variety of industrial processes and in many familiar consumer products such as glass, soap and detergents, paper, textiles, and food. (Emissions from soda ash used in glass production are reported under IPCC Source Category 2A7. Glass production is its own sub-category and historical soda ash consumption figures have been adjusted to reflect this change.) After glass manufacturing, soda ash is used primarily to manufacture many sodium-base inorganic chemicals, including sodium bicarbonate, sodium chromates, sodium phosphates, and sodium silicates (USGS 2014). Internationally, two types of soda ash are produced, natural and synthetic. The United States produces only natural soda ash and is second only to China in total soda ash production. Trona is the principal ore from which natural soda ash is made.

The United States represents about one-fourth of total world soda ash output. Only two states produce natural soda ash: Wyoming and California. Of these two states, only net emissions of CO_2 from Wyoming were calculated due to specifics regarding the production processes employed in the state.[167] Based on preliminary 2013 reported data, the estimated distribution of soda ash by end-use in 2013 (excluding glass production) was chemical production, 54 percent; soap and detergent manufacturing, 14 percent; distributors, 11 percent; flue gas desulfurization, 8 percent; other uses, 8 percent; pulp and paper production, 3 percent; and water treatment, 2 percent (USGS 2014).

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[166] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdffiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

[167] In California, soda ash is manufactured using sodium carbonate-bearing brines instead of trona ore. To extract the sodium carbonate, the complex brines are first treated with CO_2 in carbonation towers to convert the sodium carbonate into sodium bicarbonate, which then precipitates from the brine solution. The precipitated sodium bicarbonate is then calcined back into sodium carbonate. Although CO_2 is generated as a byproduct, the CO_2 is recovered and recycled for use in the carbonation stage and is not emitted. A third state, Colorado, produced soda ash until the plant was idled in 2004. The lone producer of sodium bicarbonate no longer mines trona in the state. For a brief time, sodium bicarbonate was produced using soda ash feedstocks mined in Wyoming and shipped to Colorado. Prior to 2004, because the trona was mined in Wyoming, the production numbers given by the USGS included the feedstocks mined in Wyoming and shipped to Colorado. In this way, the sodium bicarbonate production that took place in Colorado was accounted for in the Wyoming numbers.

U.S. natural soda ash is competitive in world markets because the majority of the world output of soda ash is made synthetically. Although the United States continues to be a major supplier of world soda ash, China, which surpassed the United States in soda ash production in 2003, is the world's leading producer. Despite this competition, U.S. soda ash exports are expected to increase, causing domestic production to increase slightly (USGS 2013).

In 2013, CO_2 emissions from the production of soda ash from trona were approximately 1.6 MMT CO_2 Eq. (1,610 kt). Soda ash consumption in the United States generated 1.1 MMT CO_2 Eq. (1,102 kt) in 2013. Total emissions from soda ash production and consumption in 2013 were 2.7 MMT CO_2 Eq. (2,712 kt) (see Table 4-39 and Table 4-40).

Total emissions in 2013 increased by approximately 1.5 percent from emissions in 2012, and have decreased overall by approximately 1.1 percent since 1990.

Emissions have remained relatively constant over the time series with some fluctuations since 1990. In general, these fluctuations were related to the behavior of the export market and the U.S. economy. The U.S. soda ash industry continued a trend of increased production and value in 2013 since experiencing a decline in domestic and export sales caused by adverse global economic conditions in 2009. The annual average unit value of soda ash set a record high in 2012, and soda ash exports increased as well, accounting for 55 percent of total production (USGS 2013).

Table 4-39: CO_2 Emissions from Soda Ash Production and Consumption Not Associated with Glass Manufacturing (MMT CO_2 Eq.)

Year	Production	Consumption	Total
1990	1.4	1.4	2.7
2005	1.6	1.3	2.9
2009	1.4	1.1	2.5
2010	1.5	1.1	2.6
2011	1.5	1.1	2.6
2012	1.6	1.1	2.7
2013	1.6	1.1	2.7

Note: Totals may not sum due to independent rounding.

Table 4-40: CO_2 Emissions from Soda Ash Production and Consumption Not Associated with Glass Manufacturing (kt)

Year	Production	Consumption	Total
1990	1,360	1,381	2,741
2005	1,573	1,296	2,868
2009	1,397	1,091	2,488
2010	1,471	1,141	2,612
2011	1,526	1,098	2,624
2012	1,582	1,090	2,672
2013	1,610	1,102	2,712

Note: Totals may not sum due to independent rounding.

Methodology

During the production process, trona ore is calcined in a rotary kiln and chemically transformed into a crude soda ash that requires further processing. Carbon dioxide and water are generated as byproducts of the calcination process. Carbon dioxide emissions from the calcination of trona can be estimated based on the chemical reaction shown above. Based on this formula, which is consistent with an IPCC Tier 1 approach, approximately 10.27 metric tons of trona are required to generate one metric ton of CO_2, or an emission factor of 0.097 metric tons CO_2 per metric ton trona (IPCC 2006). Thus, the 17.4 million metric tons of trona mined in 2013 for soda ash production (USGS 2014) resulted in CO_2 emissions of approximately 1.6 MMT CO_2 Eq. (1,610 kt).

Once produced, most soda ash is consumed in chemical and soap production, with minor amounts in pulp and paper, flue gas desulfurization, and water treatment (excluding soda ash consumption for glass manufacturing). As soda ash is consumed for these purposes, additional CO_2 is usually emitted. In these applications, it is assumed that one mole of carbon is released for every mole of soda ash used. Thus, approximately 0.113 metric tons of carbon (or 0.415 metric tons of CO_2) are released for every metric ton of soda ash consumed.

The activity data for trona production and soda ash consumption (see Table 4-41) between 1990 and 2013 were taken from USGS Minerals Yearbook for Soda Ash (1994 through 2013) and USGS Mineral Industry Surveys for Soda Ash (USGS 2014). Soda ash production and consumption data were collected by the USGS from voluntary surveys of the U.S. soda ash industry.

Table 4-41: Soda Ash Production and Consumption Not Associated with Glass Manufacturing (kt)

Year	Production[a]	Consumption[b]
1990	14,700	3,351
2005	17,000	3,144
2009	15,100	2,647
2010	15,900	2,768
2011	16,500	2,663
2012	17,100	2,645
2013	17,400	2,674

[a] Soda ash produced from trona ore only.

[b] Soda ash consumption is sales reported by producers which exclude imports. Historically, imported soda ash is less than 1 percent of the total U.S. consumption (Kostick 2012).

Uncertainty and Time-Series Consistency

Emission estimates from soda ash production have relatively low associated uncertainty levels in that reliable and accurate data sources are available for the emission factor and activity data. Soda ash production data was collected by the USGS from voluntary surveys. A survey request was sent to each of the five soda ash producers, all of which responded, representing 100 percent of the total production data (USGS 2014a). One source of uncertainty is the purity of the trona ore used for manufacturing soda ash. The emission factor used for this estimate assumes the ore is 100 percent pure, and likely overestimates the emissions from soda ash manufacture. The average water-soluble sodium carbonate-bicarbonate content for ore mined in Wyoming ranges from 85.5 to 93.8 percent (USGS 1995).The primary source of uncertainty, however, results from the fact that emissions from soda ash consumption are dependent upon the type of processing employed by each end-use. Specific emission factors for each end-use are not available, so a Tier 1 default emission factor is used for all end uses. Therefore, there is uncertainty surrounding the emission factors from the consumption of soda ash.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-42. Soda Ash Production and Consumption CO_2 emissions were estimated to be between 2.5 and 2.9 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below and 6 percent above the emission estimate of 2.7 MMT CO_2 Eq.

Table 4-42: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Soda Ash Production and Consumption (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Soda Ash Production and Consumption	CO_2	2.7	2.5	2.9	-7%	+6%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future Inventory reports are anticipated to estimate emissions from other uses of soda ash. To add specificity, future Inventory reports will extract soda ash consumed for other uses of carbonates from the current soda ash consumption emission estimates and include them under those sources.

In examining data from EPA's GHGRP to improve the emission estimates for Soda Ash and Consumption category, particular attention will be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[168]

4.12 Petrochemical Production (IPCC Source Category 2B8)

The production of some petrochemicals results in the release of small amounts of CH_4 and CO_2 emissions. Petrochemicals are chemicals isolated or derived from petroleum or natural gas. CO_2 emissions from the production of acrylonitrile, carbon black, ethylene, ethylene dichloride, ethylene oxide and methanol; and CH_4 emissions from the production of methanol and acrylonitrile are presented here and reported under IPCC Source Category 2B5. The petrochemical industry uses primary fossil fuels (i.e., natural gas, coal, petroleum, etc.) for non-fuel purposes in the production of carbon black and other petrochemicals. Emissions from fuels and feedstocks transferred out of the system for use in energy purposes e.g. such as indirect or direct process heat or steam production are currently accounted for in the Energy Sector.

Worldwide more than 90 percent of acrylonitrile (vinyl cyanide, C_3H_3N) is made by way of direct ammoxidation of propylene with ammonia (NH_3) and oxygen over a catalyst. This process is referred to as the SOHIO process,

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[168] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdffiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

after the Standard Oil Company of Ohio (SOHIO) (IPCC 2006). The primary use of acrylonitrile is as the raw material for the manufacture of acrylic and modacrylic fibers. Other major uses include the production of plastics (acrylonitrile-butadiene-styrene (ABS) and styrene-acrylonitrile (SAN)), nitrile rubbers, nitrile barrier resins, adiponitrile and acrylamide. All U.S. acrylonitrile facilities use the SOHIO process (AN 2014). The SOHIO process involves a fluidized bed reaction of chemical-grade propylene, ammonia, and oxygen over a catalyst. The process produces acrylonitrile as its primary product and the process yield depends on the type of catalyst used and the process configuration. The ammoxidation process also produces by-product CO_2, CO, and water from the direct oxidation of the propylene feedstock, and produces other hydrocarbons from side reactions in the ammoxidation process.

Carbon black is a black powder generated by the incomplete combustion of an aromatic petroleum- or coal-based feedstock at a high temperature. Most carbon black produced in the United States is added to rubber to impart strength and abrasion resistance, and the tire industry is by far the largest consumer. The other major use of carbon black is as a pigment. The predominant process used in the United States is the furnace black (or oil furnace) process. In the furnace black process, carbon black oil (a heavy aromatic liquid) is continuously injected into the combustion zone of a natural gas-fired furnace. Furnace heat is provided by the natural gas and a portion of the carbon black feedstock; the remaining portion of the carbon black feedstock is pyrolyzed to carbon black. The resultant CO_2 and uncombusted CH_4 emissions are released from thermal incinerators used as control devices, process dryers, and equipment leaks. Carbon black is also produced in the United States by the thermal cracking of acetylene-containing feedstocks (i.e., acetylene black process), by the thermal cracking of other hydrocarbons (i.e., thermal black process), and by the open burning of carbon black feedstock (i.e., lamp black process); each of these process are used at only one U.S. plant each (The Innovation Group 2004, EPA 2000).

Ethylene (C_2H_4) is consumed in the production processes of the plastics industry including polymers such as high, low, and linear low density polyethylene (HDPE, LDPE, LLDPE), polyvinyl chloride (PVC), ethylene dichloride, ethylene oxide, and ethylbenzene. Virtually all ethylene is produced from steam cracking of ethane, propane, butane, naphtha, gas oil, and other feedstocks. The representative chemical equation for steam cracking of ethane to ethylene is shown below:

$$C_2H_6 \rightarrow C_2H_4 + H_2$$

Small amounts of CH_4 are also generated from the steam cracking process. In addition, CO_2 and CH_4 emissions are also generated from combustion units..

Ethylene dichloride ($C_2H_4Cl_2$) is used to produce vinyl chloride monomer, which is the precursor to polyvinyl chloride (PVC). Ethylene dichloride was used as a fuel additive until 1996 when leaded gasoline was phased out. Ethylene dichloride is produced from ethylene by either direct chlorination, oxychlorination, or a combination of the two processes (i.e., the "balanced process"); most U.S. facilities use the balanced process. The direct chlorination and oxychlorination reactions are shown below:

$$C_2H_4 + Cl_2 \rightarrow C_2H_4Cl_2 \text{ (direct chlorination)}$$

$$C_2H_4 + \tfrac{1}{2}O_2 + 2HCl \rightarrow C_2H_4Cl_2 + 2H_2O \text{ (oxychlorination)}$$

$$C_2H_4 + 3O_2 \rightarrow 2CO_2 + 2H_2O \text{ (direct oxidation of ethylene during oxychlorination)}$$

In addition to the by-product CO_2 produced from the direction oxidation of the ethylene feedstock, CO_2 and CH_4 emissions are also generated from combustion units.

Ethylene oxide (C_2H_4O) is used in the manufacture of glycols, glycol ethers, alcohols, and amines. Worldwide approximately 70 percent of ethylene oxide produced is used in the manufacture of glycols, including monoethylene glycol. Ethylene oxide is produced by reacting ethylene with oxygen over a catalyst. The oxygen may be supplied to the process through either an air (air process) or a pure oxygen stream (oxygen process). The by-product CO_2 from the direct oxidation of the ethylene feedstock is removed from the process vent stream using a recycled carbonate solution, and the recovered CO_2 may be vented to the atmosphere or recovered for further utilization in other sectors, such as food production (IPCC 2006). The combined ethylene oxide reaction and by-product CO_2 reaction is exothermic and generates heat, which is recovered to produce steam for the process. The ethylene oxide process also produces other liquid and off-gas by-products (e.g., ethane) that may be burned for energy recovery within the process. Almost all facilities, except one in Texas, use the oxygen process to manufacture ethylene oxide (EPA 2008).

Methanol (CH_3OH) is a chemical feedstock most often converted into formaldehyde, acetic acid and olefins. It is also an alternative transportation fuel, as well as an additive used by municipal wastewater treatment facilities in the denitrification of wastewater. Methanol is most commonly synthesized from a synthesis gas (i.e., "syngas" – a mixture containing H_2, CO, and CO_2) using a heterogeneous catalyst. There are a number of process techniques that can be used to produce syngas. Worldwide, steam reforming of natural gas is the most common method; however, in the United States only two facilities use steam reforming of natural gas. Other syngas production processes in the United States include partial oxidation of natural gas and coal gasification.

Emissions of CO_2 and CH_4 from petrochemical production in 2013 were 26.5 MMT CO_2 Eq. (26,514 kt CO_2) and 0.1 MMT CO_2 Eq. (3 kt CH_4), respectively (see Table 4-43 and Table 4-44). Since 1990, the total CO_2 emissions from petrochemical production increased by approximately 23 percent. Methane emissions from petrochemical (methanol and acrylonitrile) production have decreased by approximately 63 percent since 1990, given declining production.

Table 4-43: CO_2 and CH_4 Emissions from Petrochemical Production (MMT CO_2 Eq.)

Year	1990	2005	2009	2010	2011	2012	2013
CO_2	21.6	28.1	23.7	27.4	26.4	26.5	26.5
CH_4	0.2	0.1	+	0.1	+	0.1	0.1
Total	**21.9**	**28.3**	**23.8**	**27.4**	**26.4**	**26.5**	**26.6**

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
+ Does not exceed 0.05 MMT CO_2 Eq.
Note: Emission totals may not add up due to rounding

Table 4-44: CO_2 and CH_4 Emissions from Petrochemical Production (kt)

Year	1990	2005	2009	2010	2011	2012	2013
CO_2	21,633	28,124	23,706	27,388	26,396	26,477	26,514
CH_4	9	6	2	2	2	3	3

Methodology

Emissions of CO_2 and CH_4 were calculated using the estimation methods provided by the *2006 IPCC Guidelines* (IPCC 2006) and country specific methods from EPA's Greenhouse Gas Reporting Program (GHGRP). The *2006 IPCC Guidelines* Tier 1 method was used to estimate CO_2 and CH_4 emissions from production of acrylonitrile and methanol, and a country specific approach similar to the IPCC Tier 2 method was used to estimate CO_2 emissions from carbon black, ethylene, ethylene oxide, and ethylene dichloride. The Tier 2 method for petrochemicals is a total feedstock carbon mass balance method used to estimate total CO_2 emissions but is not applicable for estimating CH_4 emissions. The Tier 2 mass balance is based on the assumption that all of the carbon input to the process is converted either into primary and secondary products or into CO_2. This method accounts for all the carbon as CO_2, including CH_4.

Carbon Black, Ethylene, Ethylene Dichloride and Ethylene Oxide

CO_2 emissions and national production were aggregated directly from the GHGRP data set for 2010 through 2013. In 2013, GHGRP data reported CO_2 emissions of 3,190,199 metric tons from carbon black production; 19,545,363 metric tons of CO_2 from ethylene production; 403,122 metric tons of CO_2 from ethylene dichloride production; and 1,395,936 metric tons of CO_2 from ethylene oxide production. These emissions reflect application of a country specific approach similar to the IPCC Tier 2 method and were used to estimate CO_2 emissions from the production of carbon black, ethylene, ethylene dichloride, and ethylene oxide. Since 2010, EPA's GHGRP, under Subpart X, requires all domestic producers of petrochemicals to report annual emissions and supplemental emissions information (e.g., production data) to facilitate verification of reported emissions. Under EPA's GHGRP, petrochemical production facilities are required to use either a mass balance approach or CEMS to measure and report emissions for each petrochemical process unit to estimate facility-level process CO_2 emissions. The mass balance method is used by most facilities[169] and assumes that all the carbon input is converted into primary and secondary products, byproducts, or is emitted to the atmosphere as CO_2. To apply the mass balance, facilities must measure the volume or mass of each gaseous and liquid feedstock and product, mass rate of each solid feedstock and product, and carbon content of each feedstock and product for each process unit and sum for their facility.[170] More details on the GHG calculation and monitoring methods applicable to Petrochemical facilities can be found under Subpart X (Petrochemical Production) of the regulation (40 CFR Part 98).[171]

For prior years, for these petrochemical types, an average national CO_2 emission factor was calculated based on the 2010 through 2013 GHGRP data and applied to production for earlier years in the time series (1990 through 2009) to estimate CO_2 emissions from carbon black, ethylene, ethylene dichloride, and ethylene oxide. CO_2 emission factors were derived from EPA's GHGRP by dividing annual CO_2 emissions for petrochemical type "i" with annual production for petrochemical type "i" and then averaging the derived emission factors obtained for each calendar year 2010 through 2013 (EPA GHGRP 2014). The average emission factors for each petrochemical type were applied across all prior years because petrochemical production processes in the United States have not changed significantly since 1990, though some operational efficiencies have been implemented at facilities over the time series.

The average country-specific CO_2 emission factors that were calculated from the 2010-2013 GHGRP data are as follows:

- 2.59 metric tons CO_2/metric ton carbon black produced
- 0.79 metric tons CO_2/metric ton ethylene produced
- 0.040 metric tons CO_2/metric ton ethylene dichloride produced
- 0.46 metric tons CO_2/metric ton ethylene oxide produced

Annual production data for carbon black for 1990 through 2009 were obtained from the International Carbon Black Association (Johnson 2003 and 2005 through 2010). Annual production data for ethylene and ethylene dichloride for 1990 through 2009 were obtained from the American Chemistry Council's (ACC's) *Guide to the Business of Chemistry* (ACC 2002, 2003, 2005 through 2010). Annual production data for ethylene oxide were obtained from ACC's *U.S. Chemical Industry Statistical Handbook* for 2003 through 2009 (ACC 2014a) and from ACC's Business of Chemistry for 1990 through 2002 (ACC 2014b). As noted above, annual 2010 through 2013 production data for carbon black, ethylene, ethylene dichloride, and ethylene oxide, were obtained from EPA's GHGRP (EPA GHGRP 2014).

Acrylonitrile

CO_2 and CH_4 emissions from acrylonitrile production were estimated using the Tier 1 method in the *2006 IPCC Guidelines* (IPCC 2006). Annual acrylonitrile production data were used with IPCC default Tier 1 CO_2 and CH_4 emission factors to estimate emissions for 1990 through 2013. Emission factors used to estimate acrylonitrile production emissions are as follows:

- 0.18 kg CH_4/metric ton acrylonitrile produced
- 1.00 metric tons CO_2/metric ton acrylonitrile produced

Annual acrylonitrile production data for 1990 through 2013 were obtained from ACC's Business of Chemistry (ACC 2014b).

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[169] A few facilities producing Ethylene Dichloride used CO_2 CEMS, which has been included in the aggregated GHGRP emissions.

[170] For ethylene processes only, because nearly all process emissions are from the combustion of process off-gas. Under GHGRP, Subpart X, ethylene facilities can report emissions from burning of process gases using the optional combustion methodology for ethylene production processes, which is requires estimating emissions based on fuel quantity and carbon contents of the fuel. This is consistent with the *2006 IPCC Guidelines* (p. 3.57) which recommends including combustion emissions from fuels obtained from feedstocks (e.g. off gases) in petrochemical production under in the IPPU sector.

[171] Available online at: http://www.ecfr.gov/cgi-bin/text-idx?tpl=/ecfrbrowse/Title40/40cfr98_main_02.tpl>

Methanol

CO_2 and CH_4 emissions from methanol production were estimated using Tier 1 method in the *2006 IPCC Guidelines* (IPCC 2006). Annual methanol production data were used with IPCC default Tier 1 CO_2 and CH_4 emission factors to estimate emissions for 1990 through 2013. Emission factors used to estimate methanol production emissions are as follows:

- 2.3 kg CH_4/metric ton methanol
- 0.67 metric tons CO_2/metric ton methanol

Annual methanol production data for 1990 through 2007 were obtained from the ACC Guide to the Business of Chemistry (ACC 2002, 2003, 2005 through 2011). The ACC discontinued its data series for methanol after 2007, so methanol production data for 2008 were obtained through the Methanol Institute (Jordan 2011). Methanol production data for 2009 through 2013 were obtained from Argus Media Inc. (Argus JJ&A 2014). ACC discontinued publication of this data due to confidentiality concerns given the small number of facilities producing methanol in the United States.

Table 4-45: Production of Selected Petrochemicals (kt)

Chemical	1990	2005	2009	2010	2011	2012	2013
Carbon Black	1,307	1,651	1,080	1,309	1,338	1,283	1,228
Ethylene	16,542	23,975	22,610	24,355	25,143	24,763	25,341
Ethylene Dichloride	6,283	11,260	8,120	8,149	8,621	11,309	11,462
Ethylene Oxide	2,429	3,220	2,580	2,925	3,014	3,106	3,148
Acrylonitrile	1,215	1,325	925	1,270	1,135	1,220	1,075
Methanol	3,785	2,336	790	778	685	1,015	1,350

Uncertainty and Time-Series Consistency

The CH_4 and CO_2 emission factors used for acrylonitrile and methanol production are based on a limited number of studies. Using plant-specific factors instead of default or average factors could increase the accuracy of the emission estimates; however, such data were not available for the current publication.

The results of the quantitative uncertainty analysis for the CO_2 emissions from carbon black production, ethylene, ethylene dichloride, and ethylene oxide are based on reported GHGRP data. Refer to the methodology section for more details on how these emissions were calculated and reported to EPA's GHGRP. There is some uncertainty in the applicability of the average emission factors for each petrochemical type across all prior years. While petrochemical production processes in the United States have not changed significantly since 1990, some operational efficiencies have been implemented at facilities over the time series. The uncertainty estimates for national methanol production quantity were obtained from Argus (Argus JJ&A 2014).

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-46. Petrochemical production CO_2 emissions were estimated to be between 25.3 and 27.7 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 5 percent below to 5 percent above the emission estimate of 26.5 MMT CO_2 Eq. Petrochemical production CH_4 emissions were estimated to be between 0.03 and 0.10 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 54 percent below to 44 percent above the emission estimate of 0.08 MMT CO_2 Eq.

Table 4-46: Approach 2 Quantitative Uncertainty Estimates for CH_4 Emissions from Petrochemical Production and CO_2 Emissions from Carbon Black Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a]			
			(MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Petrochemical Production	CO_2	26.5	25.3	27.7	-5%	+5%
Petrochemical Production	CH_4	0.08	0.03	0.10	-54%	+44%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculation Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in emissions from CH_4, HFCs, PFCs, SF_6, and NF_3. The GWP of N_2O has decreased, leading to a decrease in emissions. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

In addition, methodological recalculations were applied to the entire time series to ensure time series consistency. As noted above, emission information from EPA's GHGRP were used to update estimates. Average country-specific CO_2 emission factors were derived from the 2010 through 2013 GHGRP data for carbon black, ethylene, ethylene dichloride, and ethylene oxide. Annual production and CO_2 emission factor data were obtained from EPA's GHGRP for 2010 through 2013, and were used to estimate emissions for 2010 through 2013. An average CO_2 emission factor was calculated from the 2010 through 2013 GHGRP data and was used to estimate emissions for 1990 through 2009 for carbon black, ethylene, ethylene dichloride, and ethylene oxide using historic production data compiled for 1990 through 2009 (ACC 2014a; ACC 2014b).

Note, ethylene oxide is included in the IPCC petrochemical production source category but had not been included in previous versions of this Inventory due to lack of publicly-available data. Similarly, acrylonitrile is included in the IPCC Petrochemical Production source category but had not been included in the previous Inventory due to lack of publicly-available data. Annual acrylonitrile production data for 1990 through 2013 was obtained from ACC (ACC 2014b). CO_2 and CH_4 emissions from acrylonitrile were estimated using the IPCC default Tier 1 emission factors and annual acrylonitrile production.

For the previous Inventory, only CH_4 emissions were estimated for methanol using the IPCC default Tier 1 emission factor. For the current Inventory, CO_2 emissions were also estimated for methanol using the IPCC default Tier 1 CO_2 emission factor. In the current version of the Inventory, updated methanol production data were made available through Argus (Argus JJ&A 2014) for the years 2009 through 2012. This update reflected in a decrease of CH_4 emissions from Methanol production.

Planned Improvements

Pending resources, a potential improvement to the inventory estimates for this source category would focus on analyzing the fuel and feedstock data from EPA's GHGRP to better disaggregate energy related emissions and allocate them more accurately between the Energy and IPPU sectors of the Inventory. Some degree of double counting may occur between CO_2 estimates of non-energy use of fuels in the energy sector and CO_2 process emissions from petrochemical production in this sector. Data integration is not feasible at this time as feedstock data from EIA used to estimate non-energy uses of fuels are aggregated by fuel type, rather than disaggregated by both fuel type and particular industries (e.g., petrochemical production). EPA, through GHGRP, currently does not collect complete data on quantities of fuel consumed as feedstocks by petrochemical producers, only feedstock type. Updates to reporting requirements may address this issue future reporting years for the GHGRP data allowing for easier data integration between the non-energy uses of fuels category and the petrochemicals category presented in this chapter.

4.13 HCFC-22 Production (IPCC Source Category 2B9a)

Trifluoromethane (HFC-23 or CHF_3) is generated as a byproduct during the manufacture of chlorodifluoromethane (HCFC-22), which is primarily employed in refrigeration and air conditioning systems and as a chemical feedstock for manufacturing synthetic polymers. Between 1990 and 2000, U.S. production of HCFC-22 increased significantly as HCFC-22 replaced chlorofluorocarbons (CFCs) in many applications. Between 2000 and 2007, U.S. production fluctuated but generally remained above 1990 levels. In 2008 and 2009, U.S. production declined markedly and has remained near 2009 levels since. Because HCFC-22 depletes stratospheric ozone, its production for non-feedstock uses is scheduled to be phased out by 2020 under the U.S. Clean Air Act.[172] Feedstock production, however, is permitted to continue indefinitely.

HCFC-22 is produced by the reaction of chloroform ($CHCl_3$) and hydrogen fluoride (HF) in the presence of a catalyst, $SbCl_5$. The reaction of the catalyst and HF produces $SbCl_xF_y$, (where x + y = 5), which reacts with chlorinated hydrocarbons to replace chlorine atoms with fluorine. The HF and chloroform are introduced by submerged piping into a continuous-flow reactor that contains the catalyst in a hydrocarbon mixture of chloroform and partially fluorinated intermediates. The vapors leaving the reactor contain HCFC-21 ($CHCl_2F$), HCFC-22 ($CHClF_2$), HFC-23 (CHF_3), HCl, chloroform, and HF. The under-fluorinated intermediates (HCFC-21) and chloroform are then condensed and returned to the reactor, along with residual catalyst, to undergo further fluorination. The final vapors leaving the condenser are primarily HCFC-22, HFC-23, HCl and residual HF. The HCl is recovered as a useful byproduct, and the HF is removed. Once separated from HCFC-22, the HFC-23 may be released to the atmosphere, recaptured for use in a limited number of applications, or destroyed.

Two facilities produced HCFC-22 in the U.S. in 2013. Emissions of HFC-23 from this activity in 2013 were estimated to be 4.1 MMT CO_2 Eq. (0.3 kt) (see Table 4-47). This quantity represents a 25 percent decrease from 2012 emissions and a 91 percent decline from 1990 emissions. The decrease from 2012 emissions and the decrease from 1990 emissions were caused by a decrease in HCFC-22 production and a decrease in the HFC-23 emission rate (kg HFC-23 emitted/kg HCFC-22 produced). The decrease in the emission rate is primarily attributable to six factors: (a) five plants that did not capture and destroy the HFC-23 generated have ceased production of HCFC-22 since 1990, (b) one plant that captures and destroys the HFC-23 generated began to produce HCFC-22, (c) one plant implemented and documented a process change that reduced the amount of HFC-23 generated, and (d) the same plant began recovering HFC-23, primarily for destruction and secondarily for sale, (e) another plant began destroying HFC-23, and (f) the same plant, whose emission factor was higher than that of the other two plants, ceased production of HCFC-22 in 2013.

Table 4-47: HFC-23 Emissions from HCFC-22 Production (MMT CO_2 Eq. and kt HFC-23)

Year	MMT CO_2 Eq.	kt HFC-23
1990	46.1	3
2005	20.0	1
2009	6.8	0.5
2010	8.0	0.5
2011	8.8	0.6
2012	5.5	0.4
2013	4.1	0.3

Note: Emission values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values

Methodology

To estimate HFC-23 emissions for five of the eight HCFC-22 plants that have operated in the United States since 1990, methods comparable to the Tier 3 methods in the *2006 IPCC Guidelines* (IPCC 2006) were used. Emissions for 2010 through 2013 were obtained through reports submitted by U.S. HCFC-22 production facilities to EPA's GHGRP. EPA's GHGRP mandates that all HCFC-22 production facilities report their annual emissions of HFC-23 from HCFC-22 production processes and HFC-23 destruction processes. Previously, data were obtained by EPA through collaboration with an industry association that received voluntarily reported HCFC-22 production and HFC-23 emissions annually from all U.S. HCFC-22 producers from 1990 through 2009. These emissions were aggregated and reported to EPA on an annual basis.

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[172] As construed, interpreted, and applied in the terms and conditions of the *Montreal Protocol on Substances that Deplete the Ozone Layer*. [42 U.S.C. §7671m(b), CAA §614]

For the other three plants, the last of which closed in 1993, methods comparable to the Tier 1 method in the *2006 IPCC Guidelines* were used. Emissions from these three plants have been calculated using the recommended emission factor for unoptimized plants operating before 1995 (0.04 kg HCFC-23/kg HCFC-22 produced).

The five plants that have operated since 1994 measure (or, for the plants that have since closed, measured) concentrations of HFC-23 to estimate their emissions of HFC-23. Plants using thermal oxidation to abate their HFC-23 emissions monitor the performance of their oxidizers to verify that the HFC-23 is almost completely destroyed. Plants that release (or historically have released) some of their byproduct HFC-23 periodically measure HFC-23 concentrations in the output stream using gas chromatography. This information is combined with information on quantities of products (e.g., HCFC-22) to estimate HFC-23 emissions.

To estimate 1990 through 2009 emissions, reports from an industry association were used that aggregated HCFC-22 production and HFC-23 emissions from all U.S. HCFC-22 producers and reported them to EPA (ARAP 1997, 1999, 2000, 2001, 2002, 2003, 2004, 2005, 2006, 2007, 2008, 2009, 2010). To estimate 2010 through 2013 emissions, facility-level data (including both HCFC-22 production and HFC-23 emissions) reported through the EPA's GHGRP were analyzed. In 1997 and 2008, comprehensive reviews of plant-level estimates of HFC-23 emissions and HCFC-22 production were performed (RTI 1997; RTI 2008). The 1997 and 2008 reviews enabled U.S. totals to be reviewed, updated, and where necessary, corrected, and also for plant-level uncertainty analyses (Monte-Carlo simulations) to be performed for 1990, 1995, 2000, 2005, and 2006. Estimates of annual U.S. HCFC-22 production are presented in Table 4-48.

Table 4-48: HCFC-22 Production (kt)

Year	kt
1990	139
2005	156
2009	91
2010	101
2011	110
2012	96
2013	C

Note: HCFC-22 production in 2013 is considered Confidential Business Information (CBI) as there were only two producers of HCFC-22 in 2013.

Uncertainty and Time-Series Consistency

The uncertainty analysis presented in this section was based on a plant-level Monte Carlo Stochastic Simulation for 2006. The Monte Carlo analysis used estimates of the uncertainties in the individual variables in each plant's estimating procedure. This analysis was based on the generation of 10,000 random samples of model inputs from the probability density functions for each input. A normal probability density function was assumed for all measurements and biases except the equipment leak estimates for one plant; a log-normal probability density function was used for this plant's equipment leak estimates. The simulation for 2006 yielded a 95-percent confidence interval for U.S. emissions of 6.8 percent below to 9.6 percent above the reported total.

The relative errors yielded by the Monte Carlo Stochastic Simulation for 2006 were applied to the U.S. emission estimate for 2013. The resulting estimates of absolute uncertainty are likely to be reasonably accurate because (1) the methods used by the three plants to estimate their emissions are not believed to have changed significantly since 2006, and (2) although the distribution of emissions among the plants may have changed between 2006 and 2013 (because both HCFC-22 production and the HFC-23 emission rate declined significantly), the two plants that contribute significantly to emissions were estimated to have similar relative uncertainties in their 2006 (as well as 2005) emission estimates. Thus, changes in the relative contributions of these two plants to total emissions are not likely to have a large impact on the uncertainty of the national emission estimate.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-49. HFC-23 emissions from HCFC-22 production were estimated to be between 3.8 and 4.5 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 7 percent below and 10 percent above the emission estimate of 4.1 MMT CO_2 Eq.

Table 4-49: Approach 2 Quantitative Uncertainty Estimates for HFC-23 Emissions from HCFC-22 Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
HCFC-22 Production	HFC-23	4.1	3.8	4.5	-7%	+10%

[a] Range of emissions reflects a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories), which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWP of HFC-23 has increased, leading to an overall increase in emissions. For more information please see the Recalculations and Improvements Chapter.

4.14 Carbon Dioxide Consumption (IPCC Source Category 2B10)

CO_2 is used for a variety of commercial applications, including food processing, chemical production, carbonated beverage production, and refrigeration, and is also used in petroleum production for enhanced oil recovery (EOR). Carbon dioxide used for EOR is injected into the underground reservoirs to increase the reservoir pressure to enable additional petroleum to be produced. For the most part, CO_2 used in non-EOR applications will eventually be released to the atmosphere, and for the purposes of this analysis CO_2 used in commercial applications other than EOR is assumed to be emitted to the atmosphere. Carbon dioxide used in EOR applications is discussed in the Energy Chapter under “Carbon Capture and Storage, including Enhanced Oil Recovery” and is not discussed in this section.

CO_2 is produced from naturally occurring CO_2 reservoirs, as a byproduct from the energy and industrial production processes (e.g., ammonia production, fossil fuel combustion, ethanol production), and as a byproduct from the production of crude oil and natural gas, which contain naturally occurring CO_2 as a component. Only CO_2 produced from naturally occurring CO_2 reservoirs and used in industrial applications other than EOR is included in this analysis. Neither byproduct CO_2 generated from energy nor industrial production processes nor CO_2 separated from crude oil and natural gas are included in this analysis for a number of reasons. Carbon dioxide captured from biogenic sources (e.g., ethanol production plants) is not included in the inventory. Carbon dioxide captured from crude oil and gas production is used in EOR applications and is therefore reported in the Energy Chapter. Any CO_2 captured from industrial or energy production processes (e.g., ammonia plants, fossil fuel combustion) and used in non-EOR applications is assumed to be emitted to the atmosphere. The CO_2 emissions from such capture and use are therefore accounted for under Ammonia Production, Fossil Fuel Combustion, or other appropriate source category.[173]

CO_2 is produced as a byproduct of crude oil and natural gas production. This CO_2 is separated from the crude oil and natural gas using gas processing equipment, and may be emitted directly to the atmosphere, or captured and reinjected into underground formations, used for EOR, or sold for other commercial uses. A further discussion of CO_2 used in EOR is described in the Energy Chapter under the text box titled "Carbon Dioxide Transport, Injection, and Geological Storage." The only CO_2 consumption that is accounted for in this analysis is CO_2 produced from naturally-occurring CO_2 reservoirs that is used in commercial applications other than EOR.

There are currently three facilities, one in Mississippi (Jackson Dome) and two in New Mexico (Bravo Dome and West Bravo Dome), producing CO_2 from naturally-occurring CO_2 reservoirs for use in both EOR and in other commercial applications (e.g., chemical manufacturing, food production). A fourth facility in Colorado (McCallum Dome**)** is producing CO_2 from naturally occurring CO_2 reservoirs for commercial applications only (New Mexico Bureau of Geology and Mineral Resources 2006). There are other naturally-occurring CO_2 reservoirs, mostly located in the western United States, that produce CO_2, but they are only producing CO_2 for EOR applications, not for other commercial applications (Allis et al. 2000). Carbon dioxide production from these facilities is discussed in the Energy Chapter.

In 2013, the amount of CO_2 produced by the Colorado, Mississippi, and New Mexico facilities for commercial applications and subsequently emitted to the atmosphere was 0.9 MMT CO_2 Eq. (903 kt) (see Table 4-50). This is an increase of 7 percent from the previous year and a decrease of 39 percent since 1990.

Table 4-50: CO_2 Emissions from CO_2 Consumption (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.5	1,472
2005	1.4	1,375
2009	1.8	1,795
2010	1.2	1,206
2011	0.8	802
2012	0.8	841
2013	0.9	903

Methodology

CO_2 emission estimates for 1990 through 2013 were based on production data for the four facilities currently producing CO_2 from naturally-occurring CO_2 reservoirs for use in non-EOR applications. Some of the CO_2 produced by these facilities is used for EOR and some is used in other commercial applications (e.g., chemical manufacturing, food production). It is assumed that 100 percent of the CO_2 production used in commercial applications other than EOR is eventually released into the atmosphere.

CO_2 production data and the percentage of production that was used for non-EOR applications for the Jackson Dome, Mississippi facility were obtained from Advanced Resources International (ARI 2006, 2007) for 1990 to 2000, from the Annual Reports of Denbury Resources (Denbury Resources 2002 through 2010) for 2001 to 2009, and from EPA's GHGRP data for 2010 through 2013 (EPA 2014) (see Table 4-51). Denbury Resources reported the average CO_2 production in units of MMCF CO_2 per day for 2001 through 2009 and reported the percentage of the total average annual production that was used for EOR. Production from 1990 to 1999 was set equal to 2000 production, due to lack of publicly available production data for 1990-1999. Carbon dioxide production data for the Bravo Dome, New Mexico facilities were obtained from ARI for 1990 through 2009 (ARI 1990-2010), and from

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[173] There are currently four known electric power plants operating in the United States that capture CO_2 for use as food-grade CO_2 or other industrial processes; however, insufficient data prevents estimating emissions from these activities as part of CO_2 Consumption.

EPA's GHGRP data for 2010 through 2013 (EPA 2014). Data for the West Bravo Dome facility were only available starting 2009 (i.e., only for 2009 through 2013). The percentage of total production that was used for non-EOR applications for 2010 through 2013 was obtained from EPA's GHGRP (EPA 2014) data. The percentage of total production that was used for non-EOR applications for the Bravo Dome facilities for 1990 through 2009 were obtained from New Mexico Bureau of Geology and Mineral Resources (Broadhead 2003 and New Mexico Bureau of Geology and Mineral Resources 2006). Production data for the McCallum Dome (Jackson County), Colorado facility were obtained from the Colorado Oil and Gas Conservation Commission (COGCC) for 1999 through 2013 (COGCC 2014). Production data for 1990 to 1998 and percentage of production used for EOR were assumed to be the same as for 1999, due to lack of publicly-available data.

Table 4-51: CO_2 Production (kt CO_2) and the Percent Used for Non-EOR Applications

Year	Jackson Dome, MS CO_2 Production (kt) (% Non-EOR)	Bravo Dome, NM CO_2 Production (kt) (% Non-EOR)	West Bravo Dome, NM CO_2 Production (kt) (% Non-EOR)	McCallum Dome, CO CO_2 Production (kt) (% Non-EOR)
1990	1,344 (100%)	63 (1%)	+	65 (100%)
2005	1,254 (27%)	58 (1%)	+	63 (100%)
2009	1,705 (13%)	46 (1%)	21 (1%)	23 (100%)
2010	1,156 (21%)	+	+	50 (100%)
2011	770 (15%)	+	+	32 (100%)
2012	808 (16%)	+	+	33 (100%)
2013	891[174]	+	+	12 (100%)

+ Does not exceed 0%.

Uncertainty and Time-Series Consistency

Uncertainty is associated with the number of facilities that are currently producing CO_2 from naturally occurring CO_2 reservoirs for commercial uses other than EOR, and for which the CO_2 emissions are not accounted for elsewhere. Research indicates that there are only two such facilities, which are in New Mexico and Mississippi; however, additional facilities may exist that have not been identified. In addition, it is possible that CO_2 recovery exists in particular production and end-use sectors that are not accounted for elsewhere. Such recovery may or may not affect the overall estimate of CO_2 emissions from that sector depending upon the end use to which the recovered CO_2 is applied. Further research is required to determine whether CO_2 is being recovered from other facilities for application to end uses that are not accounted for elsewhere.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-52. Carbon dioxide consumption CO_2 emissions for 2013 were estimated to be between 0.8 and 1.1 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below to 13 percent above the emission estimate of 0.9 MMT CO_2 Eq.

Table 4-52: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from CO_2 Consumption (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a]			
			(MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
CO_2 Consumption	CO_2	0.9	0.8	1.1	-12%	+13%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

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[174] CO_2 quantity used for EOR applications is not yet available. The indicated quantity (891 kt) for Jackson Dome is for non-EOR applications only.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Relative to the previous Inventory, 1990 through 2009 CO_2 consumption data for the McCallum Dome facility in Colorado was corrected after a unit conversion error was identified. The revised time-series data were double checked against data reported by the Colorado Oil and Gas Conservation Commissions (COGCC 1990-2013). This revision caused an increase in CO_2 emissions for McCallum Dome for 1990 through 2009.

Planned Improvements

CO_2 production data for 1990 through 1998 for McCallum dome needs to be compiled and improved. Currently, only 1999 through 2013 data is available online (COGCC 2014). Similarly, 1990 through 1999 production data for the Jackson Dome facility is not publicly available and needs to be compiled. For example, the information could be in hard copy records at the Oil and Gas Conservation Commission and a request or site visit is required to gather the data.

4.15 Phosphoric Acid Production (IPCC Source Category 2B10)

Phosphoric acid (H_3PO_4) is a basic raw material used in the production of phosphate-based fertilizers. Phosphoric acid production from natural phosphate rock is a source of CO_2 emissions, due to the chemical reaction of the inorganic carbon (calcium carbonate) component of the phosphate rock.

Phosphate rock is mined in Florida, North Carolina, Idaho, Utah, and other areas of the United States and is used primarily as a raw material for phosphoric acid production.

The composition of natural phosphate rock varies depending upon the location where it is mined. Natural phosphate rock mined in the United States generally contains inorganic carbon in the form of calcium carbonate (limestone) and also may contain organic carbon.

The calcium carbonate component of the phosphate rock is integral to the phosphate rock chemistry. Phosphate rock can also contain organic carbon that is physically incorporated into the mined rock but is not an integral component of the phosphate rock chemistry.

The phosphoric acid production process involves chemical reaction of the calcium phosphate ($Ca_3(PO_4)_2$) component of the phosphate rock with sulfuric acid (H_2SO_4) and recirculated phosphoric acid (H_3PO_4) (EFMA 2000). However, the generation of CO_2 is due to the associated limestone-sulfuric acid reaction, as shown below:

$$CaCO_3 + H_2SO_4 + H_2O \rightarrow CaSO_4 \bullet 2H_2O + CO_2$$

Total U.S. phosphate rock production sold or used in 2013 was 29.0 million metric tons (USGS 2014). Approximately 80 percent of domestic phosphate rock production was mined in Florida and North Carolina (8 mines total), while the remaining 20 percent of production was mined in Idaho and Utah (5 mines total). Total imports of phosphate rock in 2013 were 2.6 million metric tons (USGS 2014). Most of the imported phosphate rock (70 percent) is from Morocco, with the remaining 30 percent being from Peru (USGS 2014). All phosphate rock mining companies are vertically integrated with fertilizer plants that produce phosphoric acid located near the mines. Some additional phosphoric acid production facilities are located in Texas, Louisiana, and Mississippi that used imported phosphate rock.

Over the 1990 to 2013 period, domestic production has decreased by nearly 42 percent. Total CO_2 emissions from phosphoric acid production were 1.2 MMT CO_2 Eq. (1,173 kt) in 2013 (see Table 4-53). Domestic consumption of phosphate rock in 2013 was estimated to have increased by approximately 4 percent over 2012 levels, owing to increased production of phosphoric acid (USGS 2014).

Table 4-53: CO_2 Emissions from Phosphoric Acid Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	1.6	1,586
2005	1.4	1,395
2009	1.0	1,016
2010	1.1	1,130
2011	1.2	1,198
2012	1.1	1,138
2013	1.2	1,173

Methodology

CO_2 emissions from production of phosphoric acid from phosphate rock are estimated by multiplying the average amount of inorganic carbon (expressed as CO_2) contained in the natural phosphate rock as calcium carbonate by the amount of phosphate rock that is used annually to produce phosphoric acid, accounting for domestic production and net imports for consumption. The estimation methodology is as follows:

$$E_{pa} = C_{pr} \times Q_{pr}$$

where,

E_{pa} = CO_2 emissions from phosphoric acid production, metric tons
C_{pr} = Average amount of carbon (expressed as CO_2) in natural phosphate rock, metric ton CO_2/ metric ton phosphate rock
Q_{pr} = Quantity of phosphate rock used to produce phosphoric acid

The CO_2 emissions calculation methodology is based on the assumption that all of the inorganic carbon (calcium carbonate) content of the phosphate rock reacts to CO_2 in the phosphoric acid production process and is emitted with the stack gas. The methodology also assumes that none of the organic carbon content of the phosphate rock is converted to CO_2 and that all of the organic carbon content remains in the phosphoric acid product.

From 1993 to 2004, the *USGS Mineral Yearbook: Phosphate Rock* disaggregated phosphate rock mined annually in Florida and North Carolina from phosphate rock mined annually in Idaho and Utah, and reported the annual amounts of phosphate rock exported and imported for consumption (see Table 4-54). For the years 1990 through 1992, and 2005 through 2013, only nationally aggregated mining data was reported by USGS. For the years 1990, 1991, and 1992, the breakdown of phosphate rock mined in Florida and North Carolina, and the amount mined in Idaho and Utah, are approximated using average share of U.S. production in those states from 1993 to 2004 data. For the years 2005 through 2013, the same approximation method is used, but the share of U.S. production in those states data were obtained from the USGS commodity specialist for phosphate rock (USGS 2012). Data for domestic sales or consumption of phosphate rock, exports of phosphate rock (primarily from Florida and North Carolina), and imports of phosphate rock for consumption for 1990 through 2013 were obtained from *USGS Minerals Yearbook: Phosphate Rock* (USGS 1994 through 2013), and from *USGS Minerals Commodity Summaries: Phosphate Rock in 2013* (USGS 2014). From 2004 through 2013, the USGS reported no exports of phosphate rock from U.S. producers (USGS 2005 through 2014).

The carbonate content of phosphate rock varies depending upon where the material is mined. Composition data for domestically mined and imported phosphate rock were provided by the Florida Institute of Phosphate Research (FIPR 2003). Phosphate rock mined in Florida contains approximately 1 percent inorganic carbon, and phosphate rock imported from Morocco contains approximately 1.46 percent inorganic carbon. Calcined phosphate rock mined in North Carolina and Idaho contains approximately 0.41 percent and 0.27 percent inorganic carbon, respectively (see Table 4-55).

Carbonate content data for phosphate rock mined in Florida are used to calculate the CO_2 emissions from consumption of phosphate rock mined in Florida and North Carolina (80 percent of domestic production) and carbonate content data for phosphate rock mined in Morocco are used to calculate CO_2 emissions from consumption of imported phosphate rock. The CO_2 emissions calculation is based on the assumption that all of the domestic production of phosphate rock is used in uncalcined form. As of 2006, the USGS noted that one phosphate rock producer in Idaho produces calcined phosphate rock; however, no production data were available for this single producer (USGS 2006). The USGS confirmed that no significant quantity of domestic production of phosphate rock is in the calcined form (USGS 2012b).

Table 4-54: Phosphate Rock Domestic Consumption, Exports, and Imports (kt)

Location/Year	1990	2005	2009	2010	2011	2012	2013
U.S. Domestic Consumption[a]	49,800	35,200	25,500	28,100	28,600	27,300	29,000
FL and NC	42,494	28,160	20,400	22,480	22,880	21,840	23,200
ID and UT	7,306	7,040	5,100	5,620	5,720	5,460	5,800
Exports—FL and NC	6,240	+	+	+	+	+	+
Imports	451	2,630	2,000	2,400	3,350	3,080	2,600
Total U.S. Consumption	**44,011**	**37,830**	**27,500**	**30,500**	**31,950**	**30,380**	**31,600**

Table 4-55: Chemical Composition of Phosphate Rock (Percent by weight)

Composition	Central Florida	North Florida	North Carolina (calcined)	Idaho (calcined)	Morocco
Total Carbon (as C)	1.60	1.76	0.76	0.60	1.56
Inorganic Carbon (as C)	1.00	0.93	0.41	0.27	1.46
Organic Carbon (as C)	0.60	0.83	0.35	0.00	0.10
Inorganic Carbon (as CO_2)	3.67	3.43	1.50	1.00	5.00

Source: FIPR 2003

Uncertainty and Time-Series Consistency

Phosphate rock production data used in the emission calculations were developed by the USGS through monthly and semiannual voluntary surveys of the active phosphate rock mines during 2013. For previous years in the time series, USGS provided the data disaggregated regionally; however, beginning in 2006, only total U.S. phosphate rock production was reported. Regional production for 2013 was estimated based on regional production data from previous years and multiplied by regionally-specific emission factors. There is uncertainty associated with the degree to which the estimated 2013 regional production data represents actual production in those regions. Total U.S. phosphate rock production data are not considered to be a significant source of uncertainty because all the domestic phosphate rock producers report their annual production to the USGS. Data for exports of phosphate rock used in the emission calculation are reported by phosphate rock producers and are not considered to be a significant source of uncertainty. Data for imports for consumption are based on international trade data collected by the U.S. Census Bureau. These U.S. government economic data are not considered to be a significant source of uncertainty.

An additional source of uncertainty in the calculation of CO_2 emissions from phosphoric acid production is the carbonate composition of phosphate rock, the composition of phosphate rock varies depending upon where the material is mined, and may also vary over time. The Inventory relies on one study (FIPR 2003) of chemical composition of the phosphate rock; limited data is available beyond this study. Another source of uncertainty is the disposition of the organic carbon content of the phosphate rock. A representative of the FIPR indicated that in the phosphoric acid production process, the organic C content of the mined phosphate rock generally remains in the phosphoric acid product, which is what produces the color of the phosphoric acid product (FIPR 2003a). Organic carbon is therefore not included in the calculation of CO_2 emissions from phosphoric acid production.

A third source of uncertainty is the assumption that all domestically-produced phosphate rock is used in phosphoric acid production and used without first being calcined. Calcination of the phosphate rock would result in conversion of some of the organic C in the phosphate rock into CO_2. However, according to air permit information available to the public, at least one facility has calcining units permitted for operation (NCDENR 2013).

Finally, USGS indicated that approximately 7 percent of domestically-produced phosphate rock is used to manufacture elemental phosphorus and other phosphorus-based chemicals, rather than phosphoric acid (USGS 2006). According to USGS, there is only one domestic producer of elemental phosphorus, in Idaho, and no data were available concerning the annual production of this single producer. Elemental phosphorus is produced by reducing phosphate rock with coal coke, and it is therefore assumed that 100 percent of the carbonate content of the phosphate rock will be converted to CO_2 in the elemental phosphorus production process. The calculation for CO_2 emissions is based on the assumption that phosphate rock consumption, for purposes other than phosphoric acid production, results in CO_2 emissions from 100 percent of the inorganic carbon content in phosphate rock, but none from the organic carbon content.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-56. Phosphoric acid production CO_2 emissions were estimated to be between 1.0 and 1.4 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 19 percent below and 21 percent above the emission estimate of 1.2 MMT CO_2 Eq.

Table 4-56: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Phosphoric Acid Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Phosphoric Acid Production	CO_2	1.2	1.0	1.4	-19%	+21%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Relative to the previous Inventory, the phosphate rock consumption data (sold or used and imports for consumption) for 2012 were revised based on updated data publicly available from USGS (2014). This revision caused an increase in the 2012 emission estimate by approximately 3 percent.

Planned Improvements

Pending resources, a potential improvement to the Inventory estimates for this source category would include direct integration of GHGRP data for 2010 through 2013 and use of reported GHGRP data to update the inorganic C content of phosphate rock for prior years. In order to provide estimates for the entire time series (i.e. 1990 through 2009), the applicability of the averaged inorganic C content data (by region) from 2010 through 2013 GHGRP data to previous years' estimates will need to be evaluated. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[175]

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[175] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

4.16 Iron and Steel Production (IPCC Source Category 2C1) and Metallurgical Coke Production

Iron and steel production is a multi-step process that generates process-related emissions of CO_2 and CH_4 as raw materials are refined into iron and then transformed into crude steel. Emissions from conventional fuels (e.g., natural gas, fuel oil, etc.) consumed for energy purposes during the production of iron and steel are accounted for in the Energy chapter.

Iron and steel production includes six distinct production processes: coke production, sinter production, direct reduced iron (DRI) production, pig iron production, electric arc furnace (EAF) steel production, and basic oxygen furnace (BOF) steel production. The number of production processes at a particular plant is dependent upon the specific plant configuration. In addition to the production processes mentioned above, CO_2 is also generated at iron and steel mills through the consumption of process byproducts (e.g., blast furnace gas, coke oven gas, etc.) used for various purposes including heating, annealing, and electricity generation. Process byproducts sold for use as synthetic natural gas are deducted and reported in the Energy chapter. In general, CO_2 emissions are generated in these production processes through the reduction and consumption of various carbon-containing inputs (e.g., ore, scrap, flux, coke byproducts, etc.). In addition, fugitive CH_4 emissions are also generated by the coke production, sinter production, and pig iron production processes.

Currently, there are between 15 and 20 integrated iron and steel steelmaking facilities that utilize BOFs to refine and produce steel from iron and more than 100 steelmaking facilities that utilize EAFs to produce steel primarily from recycled ferrous scrap. In addition, there are 18 cokemaking facilities, of which 7 facilities are co-located with integrated iron and steel facilities. Nearly 62 percent of the raw steel produced in the United States is produced in one of seven states: Alabama, Arkansas, Indiana, Kentucky, Mississippi, Ohio, and Tennessee.

Total production of crude steel in the United States between 2000 and 2008 ranged from a low of 99,320,000 tons to a high of 109,880,000 tons (2001 and 2004, respectively). Due to the decrease in demand caused by the global economic downturn (particularly from the automotive industry), crude steel production in the United States sharply decreased to 65,459,000 tons in 2009. In 2010, crude steel production rebounded to 88,731,000 tons as economic conditions improved and then continued to increase to 95,237,000 tons in 2011 and 97,770,000 tons in 2012; crude steel production slightly decreased to 95,766,000 tons in 2013 (AISI 2014a). The United States was the third largest producer of raw steel in the world, behind China and Japan, accounting for approximately 5.4 percent of world production in 2013 (AISI 2014a).

The majority of CO_2 emissions from the iron and steel production process come from the use of coke in the production of pig iron and from the consumption of other process byproducts, with lesser amounts emitted from the use of flux and from the removal of C from pig iron used to produce steel.

According to the *2006 IPCC Guidelines* (IPCC 2006), the production of metallurgical coke from coking coal is considered to be an energy use of fossil fuel and the use of coke in iron and steel production is considered to be an industrial process source. Therefore, the *2006 IPCC Guidelines* suggest that emissions from the production of metallurgical coke should be reported separately in the Energy sector, while emissions from coke consumption in iron and steel production should be reported in the IPPU sector. However, the approaches and emission estimates for both metallurgical coke production and iron and steel production are both presented here because the activity data used to estimate emissions from metallurgical coke production have significant overlap with activity data used to estimate iron and steel production emissions. In addition, some byproducts (e.g., coke oven gas, etc.) of the metallurgical coke production process are consumed during iron and steel production, and some byproducts of the iron and steel production process (e.g., blast furnace gas, etc.) are consumed during metallurgical coke production. Emissions associated with the consumption of these byproducts are attributed at the point of consumption. Emissions associated with the use of conventional fuels (e.g., natural gas, fuel oil, etc.) for electricity generation, heating and annealing, or other miscellaneous purposes downstream of the iron and steelmaking furnaces are reported in the Energy chapter.

Metallurgical Coke Production

Emissions of CO_2 and CH_4 from metallurgical coke production in 2013 were 1.8 MMT CO_2 Eq. (1,822 kt) and less than 0.05 MMT CO_2 Eq. (less than 0.5 kt), respectively (see Table 4-57 and Table 4-58), totaling 1.8 MMT CO2 Eq. Emissions increased in 2013 from 2012 levels, but have decreased overall since 1990. In 2013, domestic coke production increased by 1 percent from the previous year, and has decreased overall since 1990. Coke production in 2013 was 26 percent lower than in 2000 and 45 percent below 1990. Overall, emissions from metallurgical coke production have declined by 26 percent (0.6 MMT CO_2 Eq.) from 1990 to 2013.

Table 4-57: CO_2 and CH_4 Emissions from Metallurgical Coke Production (MMT CO_2 Eq.)

Gas	1990	2005	2009	2010	2011	2012	2013
CO_2	2.5	2.0	1.0	2.1	1.4	0.5	1.8
CH_4	+	+	+	+	+	+	+
Total	**2.5**	**2.0**	**1.0**	**2.1**	**1.4**	**0.5**	**1.8**

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-58: CO_2 and CH_4 Emissions from Metallurgical Coke Production (kt)

Gas	1990	2005	2009	2010	2011	2012	2013
CO_2	2,470	2,043	956	2,084	1,425	542	1,822
CH_4	+	+	+	+	+	+	+

+ Does not exceed 0.5 kt

Iron and Steel Production

Emissions of CO_2 and CH_4 from iron and steel production in 2013 were 50.5 MMT CO_2 Eq. (50,466 kt) and 0.7 MMT CO_2 Eq. (27.7 kt), respectively (see Table 4-59 through Table 4-62), totaling approximately 51.2 MMT CO_2 Eq. Emissions decreased in 2013 and have decreased overall since 1990 due to restructuring of the industry, technological improvements, and increased scrap steel utilization. Carbon dioxide emission estimates include emissions from the consumption of carbonaceous materials in the blast furnace, EAF, and BOF, as well as blast furnace gas and coke oven gas consumption for other activities at the steel mill.

In 2013, domestic production of pig iron decreased by 5 percent from 2012 levels. Overall, domestic pig iron production has declined since the 1990s. Pig iron production in 2013 was 37 percent lower than in 2000 and 39 percent below 1990. Carbon dioxide emissions from steel production have increased by 8 percent (0.7 MMT CO_2 Eq.) since 1990, while overall CO_2 emissions from iron and steel production have declined by 48 percent (46.8 MMT CO_2 Eq.) from 1990 to 2013.

Table 4-59: CO_2 Emissions from Iron and Steel Production (MMT CO_2 Eq.)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	2.4	1.7	0.8	1.0	1.2	1.2	1.1
Iron Production	47.6	19.4	15.9	19.1	19.9	12.6	13.4
Steel Production	8.0	9.4	7.6	9.2	9.3	9.9	8.6
Other Activities[a]	39.3	34.2	17.8	24.3	28.2	30.2	27.3
Total	**97.3**	**64.6**	**42.1**	**53.7**	**58.6**	**53.8**	**50.5**

[a] Includes emissions from blast furnace gas and coke oven gas combustion for activities at the steel mill other than consumption in blast furnace, EAFs, or BOFs.
Note: Totals may not sum due to independent rounding.

Table 4-60: CO_2 Emissions from Iron and Steel Production (kt)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	2,448	1,663	763	1,045	1,188	1,159	1,117
Iron Production	47,650	19,414	15,941	19,109	19,901	12,557	13,411

Steel Production	7,958	9,386	7,555	9,248	9,262	9,874	8,629
Other Activities [a]	39,256	34,160	17,815	24,260	28,232	30,195	27,309
Total	**97,311**	**64,623**	**42,073**	**53,662**	**58,583**	**53,786**	**50,466**

[a] Includes emissions from blast furnace gas and coke oven gas combustion for activities at the steel mill other than consumption in blast furnace, EAFs, or BOFs.

Note: Totals may not sum due to independent rounding.

Table 4-61: CH_4 Emissions from Iron and Steel Production (MMT CO_2 Eq.)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	+	+	+	+	+	+	+
Iron Production	1.1	0.8	0.4	0.6	0.7	0.7	0.7
Total	**1.1**	**0.9**	**0.4**	**0.6**	**0.7**	**0.7**	**0.7**

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-62: CH_4 Emissions from Iron and Steel Production (kt)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production	0.9	0.6	0.3	0.4	0.4	0.4	0.4
Iron Production	44.7	33.5	17.1	24.2	27.2	28.9	27.3
Total	**45.6**	**34.1**	**17.4**	**24.5**	**27.6**	**29.3**	**27.7**

Methodology

Emission estimates presented in this chapter are largely based on Tier 2 methodologies provided by the *2006 IPCC Guidelines* (IPCC 2006). These Tier 2 methodologies call for a mass balance accounting of the carbonaceous inputs and outputs during the iron and steel production process and the metallurgical coke production process. Tier 1 methods are used for certain iron and steel production processes (i.e., sinter production and DRI production) for which available data are insufficient for utilizing a Tier 2 method.

The Tier 2 methodology equation is as follows:

$$E_{CO_2} = \left[\sum_a (Q_a \times C_a) - \sum_b (Q_b \times C_b)\right] \times \frac{44}{12}$$

where,

E_{CO2} = Emissions from coke, pig iron, EAF steel, or BOF steel production, metric tons
a = Input material *a*
b = Output material *b*
Q_a = Quantity of input material *a*, metric tons
C_a = Carbon content of material *a*, metric tons C/metric ton material
Q_b = Quantity of output material *b*, metric tons
C_b = Carbon content of material *b*, metric tons C/metric ton material
44/12 = Stoichiometric ratio of CO_2 to C

The Tier 1 methodology equations are as follows:

$$E_{s,p} = Q_s \times EF_{s,p}$$

$$E_{d,p} = Q_d \times EF_{d,p}$$

where,

$E_{s,p}$ = Emissions from sinter production process for pollutant *p* (CO_2 or CH_4), metric ton

Q_s = Quantity of sinter produced, metric tons
$EF_{s,p}$ = Emission factor for pollutant p (CO_2 or CH_4), metric ton p/metric ton sinter
$E_{d,p}$ = Emissions from DRI production process for pollutant p (CO_2 or CH_4), metric ton
Q_d = Quantity of DRI produced, metric tons
$EF_{d,p}$ = Emission factor for pollutant p (CO_2 or CH_4), metric ton p/metric ton DRI

Metallurgical Coke Production

Coking coal is used to manufacture metallurgical (coal) coke that is used primarily as a reducing agent in the production of iron and steel, but is also used in the production of other metals including zinc and lead (see Zinc Production and Lead Production sections of this chapter). Emissions associated with producing metallurgical coke from coking coal are estimated and reported separately from emissions that result from the iron and steel production process. To estimate emission from metallurgical coke production, a Tier 2 method provided by the *2006 IPCC Guidelines* (IPCC 2006) was utilized. The amount of C contained in materials produced during the metallurgical coke production process (i.e., coke, coke breeze, coke oven gas, and coal tar) is deducted from the amount of carbon contained in materials consumed during the metallurgical coke production process (i.e., natural gas, blast furnace gas, and coking coal). Light oil, which is produced during the metallurgical coke production process, is excluded from the deductions due to data limitations. The amount of C contained in these materials is calculated by multiplying the material-specific carbon content by the amount of material consumed or produced (see Table 4-63). The amount of coal tar produced was approximated using a production factor of 0.03 tons of coal tar per ton of coking coal consumed. The amount of coke breeze produced was approximated using a production factor of 0.075 tons of coke breeze per ton of coking coal consumed (AISI 2008c; DOE 2000). Data on the consumption of carbonaceous materials (other than coking coal) as well as coke oven gas production were available for integrated steel mills only (i.e., steel mills with co-located coke plants). Therefore, carbonaceous material (other than coking coal) consumption and coke oven gas production were excluded from emission estimates for merchant coke plants. Carbon contained in coke oven gas used for coke-oven underfiring was not included in the deductions to avoid double-counting.

Table 4-63: Material Carbon Contents for Metallurgical Coke Production

Material	kg C/kg
Coal Tar	0.62
Coke	0.83
Coke Breeze	0.83
Coking Coal	0.73
Material	**kg C/GJ**
Coke Oven Gas	12.1
Blast Furnace Gas	70.8

Source: IPCC 2006, Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

The production processes for metallurgical coke production results in fugitive emissions of CH_4, which are emitted via leaks in the production equipment, rather than through the emission stacks or vents of the production plants. The fugitive emissions were calculated by applying Tier 1 emission factors (0.1g CH_4 per metric ton of coke production) taken from the *2006 IPCC Guidelines* (IPCC 2006) for metallurgical coke production.

Data relating to the mass of coking coal consumed at metallurgical coke plants and the mass of metallurgical coke produced at coke plants were taken from the Energy Information Administration (EIA), *Quarterly Coal Report: October through December* (EIA 1998 through 2014d) (see Table 4-64). Data on the volume of natural gas consumption, blast furnace gas consumption, and coke oven gas production for metallurgical coke production at integrated steel mills were obtained from the American Iron and Steel Institute (AISI), *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c) (see Table 4-65). The factor for the quantity of coal tar produced per ton of coking coal consumed was provided by AISI (2008c). The factor for the quantity of coke breeze produced per ton of coking coal consumed was obtained through Table 2-1 of the report, *Energy and Environmental Profile of the U.S. Iron and Steel Industry* (DOE 2000). Data on natural gas consumption and coke oven gas production at merchant coke plants were not available and were excluded from the emission estimate. Carbon contents for coking coal, metallurgical coke, coal tar, coke oven gas, and blast furnace gas were provided by the *2006 IPCC Guidelines* (IPCC 2006). The carbon content for coke breeze was assumed to equal the C content of coke.

Table 4-64: Production and Consumption Data for the Calculation of CO_2 and CH_4 Emissions from Metallurgical Coke Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Metallurgical Coke Production							
Coking Coal Consumption at Coke Plants	35,269	21,259	13,904	19,135	19,445	18,825	19,481
Coke Production at Coke Plants	25,054	15,167	10,109	13,628	13,989	13,764	13,898
Coal Breeze Production	2,645	1,594	1,043	1,435	1,458	1,412	1,461
Coal Tar Production	1,058	638	417	574	583	565	584

Table 4-65: Production and Consumption Data for the Calculation of CO_2 Emissions from Metallurgical Coke Production (million ft^3)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Metallurgical Coke Production							
Coke Oven Gas Production	250,767	114,213	66,155	95,405	109,044	113,772	108,162
Natural Gas Consumption	599	2,996	2,121	3,108	3,175	3,267	3,247
Blast Furnace Gas Consumption	24,602	4,460	2,435	3,181	3,853	4,351	4,255

Iron and Steel Production

Emissions of CO_2 from sinter production and direct reduced iron production were estimated by multiplying total national sinter production and the total national direct reduced iron production by Tier 1 CO_2 emission factors (see Table 4-66). Because estimates of sinter production and direct reduced iron production were not available, production was assumed to equal consumption.

Table 4-66: CO_2 Emission Factors for Sinter Production and Direct Reduced Iron Production

Material Produced	Metric Ton CO_2/Metric Ton
Sinter	0.2
Direct Reduced Iron	0.7

Source: IPCC 2006, Table 4.1.

To estimate emissions from pig iron production in the blast furnace, the amount of C contained in the produced pig iron and blast furnace gas were deducted from the amount of C contained in inputs (i.e., metallurgical coke, sinter, natural ore, pellets, natural gas, fuel oil, coke oven gas, and direct coal injection). The C contained in the pig iron, blast furnace gas, and blast furnace inputs was estimated by multiplying the material-specific C content by each material type (see Table 4-67). Carbon in blast furnace gas used to pre-heat the blast furnace air is combusted to form CO_2 during this process.

Emissions from steel production in EAFs were estimated by deducting the C contained in the steel produced from the C contained in the EAF anode, charge carbon, and scrap steel added to the EAF. Small amounts of C from direct reduced iron, pig iron, and flux additions to the EAFs were also included in the EAF calculation. For BOFs, estimates of C contained in BOF steel were deducted from C contained in inputs such as natural gas, coke oven gas, fluxes, and pig iron. In each case, the carbon was calculated by multiplying material-specific carbon contents by each material type (see Table 4-67). For EAFs, the amount of EAF anode consumed was approximated by multiplying total EAF steel production by the amount of EAF anode consumed per metric ton of steel produced (0.002 metric tons EAF anode per metric ton steel produced [AISI 2008c]). The amount of flux (e.g., limestone and dolomite) used during steel manufacture was deducted from the Other Process Uses of Carbonates source category to avoid double-counting.

CO_2 emissions from the consumption of blast furnace gas and coke oven gas for other activities occurring at the steel mill were estimated by multiplying the amount of these materials consumed for these purposes by the material-specific carbon content (see Table 4-67).

CO_2 emissions associated with the sinter production, direct reduced iron production, pig iron production, steel production, and other steel mill activities were summed to calculate the total CO_2 emissions from iron and steel production (see Table 4-59 and Table 4-60).

Table 4-67: Material Carbon Contents for Iron and Steel Production

Material	kg C/kg
Coke	0.83
Direct Reduced Iron	0.02
Dolomite	0.13
EAF Carbon Electrodes	0.82
EAF Charge Carbon	0.83
Limestone	0.12
Pig Iron	0.04
Steel	0.01
Material	**kg C/GJ**
Coke Oven Gas	12.1
Blast Furnace Gas	70.8

Source: IPCC 2006, Table 4.3. Coke Oven Gas and Blast Furnace Gas, Table 1.3.

The production processes for sinter and pig iron result in fugitive emissions of CH_4, which are emitted via leaks in the production equipment, rather than through the emission stacks or vents of the production plants. The fugitive emissions were calculated by applying Tier 1 emission factors taken from the *2006 IPCC Guidelines* (IPCC 2006) for sinter production and the *1995 IPCC Guidelines* (IPCC/UNEP/ OECD/IEA 1995) (see Table 4-68) for pig iron production. The production of direct reduced iron also results in emissions of CH_4 through the consumption of fossil fuels (e.g., natural gas); however, these emission estimates are excluded due to data limitations.

Table 4-68: CH_4 Emission Factors for Sinter and Pig Iron Production

Material Produced	Factor	Unit
Pig Iron	0.9	g CH_4/kg
Sinter	0.07	kg CH_4/metric ton

Source: Sinter (IPCC 2006, Table 4.2), Pig Iron (IPCC/UNEP/OECD/IEA 1995, Table 2.2)

Sinter consumption data for 1990 through 2013 were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c) (see Table 4-69). In general, direct reduced iron (DRI) consumption data were obtained from the *USGS Minerals Yearbook – Iron and Steel Scrap* (USGS 1991 through 2013) and personal communication with the USGS Iron and Steel Commodity Specialist (Fenton 2014). However, data for DRI consumed in EAFs were not available for the years 1990 and 1991. EAF DRI consumption in 1990 and 1991 was calculated by multiplying the total DRI consumption for all furnaces by the EAF share of total DRI consumption in 1992. Also, data for DRI consumed in BOFs were not available for the years 1990 through 1993. BOF DRI consumption in 1990 through 1993 was calculated by multiplying the total DRI consumption for all furnaces (excluding EAFs and cupola) by the BOF share of total DRI consumption (excluding EAFs and cupola) in 1994.

The Tier 1 CO_2 emission factors for sinter production and direct reduced iron production were obtained through the *2006 IPCC Guidelines* (IPCC 2006). Time series data for pig iron production, coke, natural gas, fuel oil, sinter, and pellets consumed in the blast furnace; pig iron production; and blast furnace gas produced at the iron and steel mill and used in the metallurgical coke ovens and other steel mill activities were obtained from AISI's Annual Statistical Report (AISI 2004 through 2014a) and through personal communications with AISI (2008c) (see Table 4-69 and Table 4-70).

Data for EAF steel production, flux, EAF charge carbon, and natural gas consumption were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2006 through 2014b and 2008c). The factor for the quantity of EAF anode consumed per ton of EAF steel produced was provided by AISI (AISI 2008c). Data for BOF steel production, flux, natural gas, natural ore, pellet sinter consumption as well as BOF steel production were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c). Data for EAF and BOF scrap steel, pig iron, and DRI consumption were obtained from the *USGS Minerals Yearbook – Iron and Steel Scrap* (USGS 1991 through 2013). Data on coke oven gas and blast furnace gas consumed at the iron and steel mill (other than in the EAF, BOF, or blast furnace) were obtained from AISI's *Annual Statistical Report* (AISI 2004 through 2014a) and through personal communications with AISI (2008c).

Data on blast furnace gas and coke oven gas sold for use as synthetic natural gas were obtained from EIA's Natural Gas Annual 2011 (EIA 2012b). Carbon contents for direct reduced iron, EAF carbon electrodes, EAF charge carbon, limestone, dolomite, pig iron, and steel were provided by the *2006 IPCC Guidelines* (IPCC 2006). The C contents for natural gas, fuel oil, and direct injection coal were obtained from EIA (2013c) and EPA (2010). Heat contents for the same fuels were obtained from EIA (1992, 2013a). Heat contents for coke oven gas and blast furnace gas were provided in Table 2-2 of the report, *Energy and Environmental Profile of the U.S. Iron and Steel Industry* (DOE 2000).

Table 4-69: Production and Consumption Data for the Calculation of CO_2 and CH_4 Emissions from Iron and Steel Production (Thousand Metric Tons)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Sinter Production							
Sinter Production	12,239	8,315	3,814	5,225	5,941	5,795	5,583
Direct Reduced Iron Production							
Direct Reduced Iron Production	516	1,303	1,165	1,441	1,582	3,530	3,350
Pig Iron Production							
Coke Consumption	24,946	13,832	8,572	10,883	11,962	9,571	9,308
Pig Iron Production	49,669	37,222	19,019	26,844	30,228	32,063	30,309
Direct Injection Coal Consumption	1,485	2,573	1,674	2,279	2,604	2,802	2,675
EAF Steel Production							
EAF Anode and Charge Carbon Consumption	67	1,127	845	1,189	1,257	1,318	1,122
Scrap Steel Consumption	42,691	46,600	43,200	47,500	50,500	50,900	47,327
Flux Consumption	319	695	476	640	726	748	771
EAF Steel Production	33,511	52,194	36,725	49,339	52,108	52,415	52,641
BOF Steel Production							
Pig Iron Consumption	47,307	34,400	25,900	31,200	31,300	31,500	29,570
Scrap Steel Consumption	14,713	11,400	7,110	9,860	8,800	8,350	7,894
Flux Consumption	576	582	318	431	454	476	454
BOF Steel Production	43,973	42,705	22,659	31,158	34,291	36,282	34,238

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

Table 4-70: Production and Consumption Data for the Calculation of CO_2 Emissions from Iron and Steel Production (million ft^3 unless otherwise specified)

Source/Activity Data	1990	2005	2009	2010	2011	2012	2013
Pig Iron Production							
Natural Gas Consumption	56,273	59,844	35,933	47,814	59,132	62,469	48,812
Fuel Oil Consumption (thousand gallons)	163,397	16,170	23,179	27,505	21,378	19,240	17,468
Coke Oven Gas Consumption	22,033	16,557	9,951	14,233	17,772	18,608	17,710
Blast Furnace Gas Production	1,439,380	1,299,980	672,486	911,180	1,063,326	1,139,578	1,026,973

EAF Steel Production							
Natural Gas Consumption	15,905	19,985	7,848	10,403	6,263	11,145	10,514
BOF Steel Production							
Coke Oven Gas Consumption	3,851	524	373	546	554	568	568
Other Activities							
Coke Oven Gas Consumption	224,883	97,132	55,831	80,626	90,718	94,596	89,884
Blast Furnace Gas Consumption	1,414,778	1,295,520	670,051	907,999	1,059,473	1,135,227	1,022,718

Uncertainty and Time-Series Consistency

The estimates of CO_2 and CH_4 emissions from metallurgical coke production are based on material production and consumption data and average carbon contents. Uncertainty is associated with the total U.S. coking coal consumption, total U.S. coke production and materials consumed during this process. Data for coking coal consumption and metallurgical coke production are from different data sources (EIA) than data for other carbonaceous materials consumed at coke plants (AISI), which does not include data for merchant coke plants. There is uncertainty associated with the fact that coal tar and coke breeze production were estimated based on coke production because coal tar and coke breeze production data were not available. Since merchant coke plant data is not included in the estimate of other carbonaceous materials consumed at coke plants, the mass balance equation for CO_2 from metallurgical coke production cannot be reasonably completed. Therefore, for the purpose of this analysis, uncertainty parameters are applied to primary data inputs to the calculation (i.e., coking coal consumption and metallurgical coke production) only.

The estimates of CO_2 emissions from iron and steel production are based on material production and consumption data and average C contents. There is uncertainty associated with the assumption that direct reduced iron and sinter consumption are equal to production. There is uncertainty associated with the assumption that all coal used for purposes other than coking coal is for direct injection coal; some of this coal may be used for electricity generation. There is also uncertainty associated with the C contents for pellets, sinter, and natural ore, which are assumed to equal the carbon contents of direct reduced iron. For EAF steel production, there is uncertainty associated with the amount of EAF anode and charge C consumed due to inconsistent data throughout the time series. Also for EAF steel production, there is uncertainty associated with the assumption that 100 percent of the natural gas attributed to "steelmaking furnaces" by AISI is process-related and nothing is combusted for energy purposes. Uncertainty is also associated with the use of process gases such as blast furnace gas and coke oven gas. Data are not available to differentiate between the use of these gases for processes at the steel mill versus for energy generation (i.e., electricity and steam generation); therefore, all consumption is attributed to iron and steel production. These data and C contents produce a relatively accurate estimate of CO_2 emissions. However, there are uncertainties associated with each.

For the purposes of the CH_4 calculation from iron and steel production it is assumed that all of the CH_4 escapes as fugitive emissions and that none of the CH_4 is captured in stacks or vents. Additionally, the CO_2 emissions calculation is not corrected by subtracting the C content of the CH_4, which means there may be a slight double counting of C as both CO_2 and CH_4.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-71 for metallurgical coke production and iron and steel production. Total CO_2 emissions from metallurgical coke production and iron and steel production were estimated to be between 43.3 and 61.2 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 17 percent below and 17 percent above the emission estimate of 52.3 MMT CO_2 Eq. Total CH_4 emissions from metallurgical coke production and iron and steel production were estimated to be between 0.5 and 0.8 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 21 percent below and 22 percent above the emission estimate of 0.7 MMT CO_2 Eq.

Table 4-71: Approach 2 Quantitative Uncertainty Estimates for CO_2 and CH_4 Emissions from Iron and Steel Production and Metallurgical Coke Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Metallurgical Coke & Iron and Steel Production	CO_2	52.3	43.3	61.2	-17%	+17%
Metallurgical Coke & Iron and Steel Production	CH_4	0.7	0.5	0.8	-21%	+22%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP to improve the emission estimates for the Iron and Steel Production source category. Particular attention would be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[176]

Additional improvements include accounting for emission estimates for the production of metallurgical coke to the Energy chapter as well as identifying the amount of carbonaceous materials, other than coking coal, consumed at merchant coke plants. Other potential improvements include identifying the amount of coal used for direct injection and the amount of coke breeze, coal tar, and light oil produced during coke production. Efforts will also be made to identify inputs for preparing Tier 2 estimates for sinter and direct reduced iron production, as well as identifying information to better characterize emissions from the use of process gases and fuels within the Energy and Industrial Processes chapters.

4.17 Ferroalloy Production (IPCC Source Category 2C2)

Carbon dioxide and CH_4 are emitted from the production of several ferroalloys. Ferroalloys are composites of iron (Fe) and other elements such as silicon (Si), manganese (Mn), and chromium (Cr). Emissions from fuels consumed for energy purposes during the production of ferroalloys are accounted for in the Energy chapter. Emissions from the production of two types of ferrosilicon (25 to 55 percent and 56 to 95 percent silicon), silicon metal (96 to 99 percent silicon), and miscellaneous alloys (32 to 65 percent silicon) have been calculated. Emissions from the production of ferrochromium and ferromanganese are not included here because of the small number of manufacturers of these materials in the United States, and therefore, government information disclosure rules prevent the publication of production data for these production facilities.

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[176] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

Similar to emissions from the production of iron and steel, CO_2 is emitted when metallurgical coke is oxidized during a high-temperature reaction with iron and the selected alloying element. Due to the strong reducing environment, CO is initially produced, and eventually oxidized to CO_2. A representative reaction equation for the production of 50 percent ferrosilicon (FeSi) is given below:

$$Fe_2O_3 + 2SiO_2 + 7C \rightarrow 2FeSi + 7CO$$

While most of the carbon contained in the process materials is released to the atmosphere as CO_2, a percentage is also released as CH_4 and other volatiles. The amount of CH_4 that is released is dependent on furnace efficiency, operation technique, and control technology.

When incorporated in alloy steels, ferroalloys are used to alter the material properties of the steel. Ferroalloys are used primarily by the iron and steel industry, and production trends closely follow that of the iron and steel industry. Fewer than 10 facilities in the United States produce ferroalloys.

Emissions of CO_2 from ferroalloy production in 2013 were 1.8 MMT CO_2 Eq. (1,785 kt) (see Table 4-72 and Table 4-73), which is a 17 percent reduction since 1990. Emissions of CH_4 from ferroalloy production in 2013 were 0.01 MMT CO_2 Eq. (0.5 kt CH_4), which is a 26 percent decrease since 1990.

Table 4-72: CO_2 and CH_4 Emissions from Ferroalloy Production (MMT CO_2 Eq.)

Gas	1990	2005	2009	2010	2011	2012	2013
CO_2	2.2	1.4	1.5	1.7	1.7	1.9	1.8
CH_4	+	+	+	+	+	+	+
Total	**2.2**	**1.4**	**1.5**	**1.7**	**1.7**	**1.7**	**1.8**

+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-73: CO_2 and CH_4 Emissions from Ferroalloy Production (kt)

Gas	1990	2005	2009	2010	2011	2012	2013
CO_2	2,152	1,392	1,469	1,663	1,735	1,903	1,785
CH_4	1	+	+	+	+	1	+

+ Does not exceed 0.5 kt

Methodology

Emissions of CO_2 and CH_4 from ferroalloy production were calculated using a Tier 1 method from the *2006 IPCC Guidelines* (IPCC 2006) by multiplying annual ferroalloy production by material-specific default emission factors provided by IPCC (2006). Default emission factors were used because country-specific emission factors are not currently available.

For ferrosilicon alloys containing 25 to 55 percent silicon and miscellaneous alloys (including primarily magnesium-ferrosilicon, but also including other silicon alloys) containing 32 to 65 percent silicon, an emission factor for 45 percent silicon was applied for CO_2 (i.e., 2.5 metric tons CO_2/metric ton of alloy produced) and an emission factor for 65 percent silicon was applied for CH_4 (i.e., 1 kg CH_4/metric ton of alloy produced). Additionally, for ferrosilicon alloys containing 56 to 95 percent silicon, an emission factor for 75 percent silicon ferrosilicon was applied for both CO_2 and CH_4 (i.e., 4 metric tons CO_2/metric ton alloy produced and 1 kg CH_4/metric ton of alloy produced, respectively). The emission factors for silicon metal equaled 5 metric tons CO_2/metric ton metal produced and 1.2 kg CH_4/metric ton metal produced. It was assumed that 100 percent of the ferroalloy production was produced using petroleum coke in an electric arc furnace process (IPCC 2006), although some ferroalloys may have been produced with coking coal, wood, other biomass, or graphite carbon inputs. The amount of petroleum coke consumed in ferroalloy production was calculated assuming that the petroleum coke used is 90 percent C and 10 percent inert material (Onder and Bagdoyan 1993).

Ferroalloy production data for 1990 through 2013 (see Table 4-74) were obtained from the USGS through the *Minerals Yearbook: Silicon* (USGS 1996 through 2013) and the *Mineral Industry Surveys: Silicon in September 2014* (USGS 2014). The following data were available from the USGS publications for the time-series:

- Ferrosilicon, 25%-55% Si: Annual production data were available from 1990-2010.

- Ferrosilicon, 56%-95% Si: Annual production data were available from 1990-2010.
- Silicon Metal: Annual production data were available from 1990-2005. The production data for 2005 were used as proxy for 2006-2010.
- Miscellaneous Alloys, 32%-65% Si: Annual production data were available from 1990-1999. Starting 2000, USGS reported miscellaneous alloys and ferrosilicon containing 25 to 55 percent silicon as a single category.

Starting with the 2011 publication, USGS reported all the ferroalloy production data as a single category (i.e., Total Silicon Materials Production). This is due to the small number of ferroalloy manufacturers in the United States and government information disclosure rules. Ferroalloy product shares developed from the 2010 production data (i.e., ferroalloy product production/total ferroalloy production) were used with the total silicon materials production quantity to estimate the production quantity by ferroalloy product type for 2011 through 2013 (USGS 2013, 2014). The composition data for petroleum coke was obtained from Onder and Bagdoyan (1993).

Table 4-74: Production of Ferroalloys (Metric Tons)

Year	Ferrosilicon 25%-55%	Ferrosilicon 56%-95%	Silicon Metal	Misc. Alloys 32-65%
1990	321,385	109,566	145,744	72,442
2005	123,000	86,100	148,000	NA
2009	123,932	104,855	148,000	NA
2010	153,000	135,000	148,000	NA
2011	159,667	140,883	154,450	NA
2012	175,108	154,507	169,385	NA
2013	164,229	144,908	158,862	NA

NA (Not Available for product type, aggregated along with ferrosilicon, 25%-55% Si)

Uncertainty and Time-Series Consistency

Annual ferroalloy production was reported by the USGS in three broad categories till the 2010 publication: ferroalloys containing 25 to 55 percent silicon (including miscellaneous alloys), ferroalloys containing 56 to 95 percent silicon, and silicon metal (through 2005 only, 2005 value used as proxy for 2005 through 2010). Starting with the 2011 minerals yearbook, USGS started reporting all the ferroalloy production under a single category: Total silicon materials production. The total silicon materials quantity was allocated across the three categories based on the 2010 production shares for the three categories. Refer to the Methodology section for further details. Additionally, production data for silvery pig iron (alloys containing less than 25 percent silicon) are not reported by the USGS to avoid disclosing proprietary company data. Emissions from this production category, therefore, were not estimated.

Also, some ferroalloys may be produced using wood or other biomass as a primary or secondary carbon source (carbonaceous reductants), information and data regarding these practices were not available. Emissions from ferroalloys produced with wood or other biomass would not be counted under this source because wood-based carbon is of biogenic origin.[177] Even though emissions from ferroalloys produced with coking coal or graphite inputs would be counted in national trends, they may be generated with varying amounts of CO_2 per unit of ferroalloy produced. The most accurate method for these estimates would be to base calculations on the amount of reducing agent used in the process, rather than the amount of ferroalloys produced. These data, however, were not available, and are also often considered confidential business information.

Emissions of CH_4 from ferroalloy production will vary depending on furnace specifics, such as type, operation technique, and control technology. Higher heating temperatures and techniques such as sprinkle charging will

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[177] Emissions and sinks of biogenic carbon are accounted for in the *Land Use, Land-Use Change, and Forestry* chapter.

reduce CH_4 emissions; however, specific furnace information was not available or included in the CH_4 emission estimates.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-75. Ferroalloy production CO_2 emissions were estimated to be between 1.6 and 2.0 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 12 percent below and 12 percent above the emission estimate of 1.8 MMT CO_2 Eq. Ferroalloy production CH_4 emissions were estimated to be between a range of approximately 12 percent below and 12 percent above the emission estimate of 0.01 MMT CO_2 Eq.

Table 4-75: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Ferroalloy Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Ferroalloy Production	CO_2	1.8	1.6	2.0	-12%	+12%
Ferroalloy Production	CH_4	+	+	+	-12%	+12%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.
+ Does not exceed 0.05 MMT CO_2 Eq.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

Starting in 2011, USGS ceased publication of ferrosilicon production data disaggregated by product type. Instead, total silicon materials production was reported for 2011 through 2013. The previous versions of the Inventory used 2010 production data (by product type) as proxy for 2011 and 2012. In this version of the Inventory, production shares by product type were developed using the 2010 production data (by product type). These ferrosilicon product shares were applied to the total ferrosilicon production quantity to estimate annual production by product type for 2011 through 2013.

Planned Improvements

According to the *2006 IPCC Guidelines* (IPCC 2006), emission factors are provided for a total of nine different ferroalloy types: four grades of ferrosilicon (FeSi) (i.e., 45, 65, 75, and 90 percent Si), two grades of ferromanganese (FeMn) (i.e., 1 and 7 percent C), silicomanganese (SiMn), ferrochromium (FeCr), and silicon metal. However, due to the small number of ferroalloy manufacturers in the United States and government information disclosure rules, the current availability of ferroalloy production data is quite limited (Tuck 2013). Additional research is being conducting to assess the feasibility of obtaining alternative activity data.

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP that would be useful to improve the emission estimates for the Ferroalloy Production source category. Particular attention would be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[178]

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[178] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

4.18 Aluminum Production (IPCC Source Category 2C3)

Aluminum is a light-weight, malleable, and corrosion-resistant metal that is used in many manufactured products, including aircraft, automobiles, bicycles, and kitchen utensils. As of recent reporting, the United States was the fourth largest producer of primary aluminum, with approximately 4 percent of the world total production (USGS 2014). The United States was also a major importer of primary aluminum. The production of primary aluminum—in addition to consuming large quantities of electricity—results in process-related emissions of CO_2 and two perfluorocarbons (PFCs): Perfluoromethane (CF_4) and perfluoroethane (C_2F_6).

CO_2 is emitted during the aluminum smelting process when alumina (aluminum oxide, Al_2O_3) is reduced to aluminum using the Hall-Heroult reduction process. The reduction of the alumina occurs through electrolysis in a molten bath of natural or synthetic cryolite (Na_3AlF_6). The reduction cells contain a carbon lining that serves as the cathode. Carbon is also contained in the anode, which can be a C mass of paste, coke briquettes, or prebaked C blocks from petroleum coke. During reduction, most of this C is oxidized and released to the atmosphere as CO_2.

Process emissions of CO_2 from aluminum production were estimated to be 3.3 MMT CO_2 Eq. (3,255 kt) in 2013 (see Table 4-76). The C anodes consumed during aluminum production consist of petroleum coke and, to a minor extent, coal tar pitch. The petroleum coke portion of the total CO_2 process emissions from aluminum production is considered to be a non-energy use of petroleum coke, and is accounted for here and not under the CO_2 from Fossil Fuel Combustion source category of the Energy sector. Similarly, the coal tar pitch portion of these CO_2 process emissions is accounted for here.

Table 4-76: CO_2 Emissions from Aluminum Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	6.8	6,831
2005	4.1	4,142
2009	3.0	3,009
2010	2.7	2,722
2011	3.3	3,292
2012	3.4	3,439
2013	3.3	3,255

In addition to CO_2 emissions, the aluminum production industry is also a source of PFC emissions. During the smelting process, when the alumina ore content of the electrolytic bath falls below critical levels required for electrolysis, rapid voltage increases occur, which are termed "anode effects." These anode effects cause C from the anode and fluorine from the dissociated molten cryolite bath to combine, thereby producing fugitive emissions of CF_4 and C_2F_6. In general, the magnitude of emissions for a given smelter and level of production depends on the frequency and duration of these anode effects. As the frequency and duration of the anode effects increase, emissions increase.

Since 1990, emissions of CF_4 and C_2F_6 have declined by 87 percent and 81 percent, respectively, to 2.3 MMT CO_2 Eq. of CF_4 (0.31 kt) and 0.7 MMT CO_2 Eq. of C_2F_6 (0.05 kt) in 2013, as shown in Table 4-77 and Table 4-78. This decline is due both to reductions in domestic aluminum production and to actions taken by aluminum smelting companies to reduce the frequency and duration of anode effects. These actions include technology and operational changes such as employee training, use of computer monitoring, and changes in alumina feeding techniques. Since 1990, aluminum production has declined by 52 percent, while the combined CF_4 and C_2F_6 emission rate (per metric ton of aluminum produced) has been reduced by 71 percent. Emissions increased by approximately 1 percent between 2012 and 2013 due to a slight increase in both CF_4 and C_2F_6 emissions per metric ton of aluminum produced.

Table 4-77: PFC Emissions from Aluminum Production (MMT CO_2 Eq.)

Year	CF_4	C_2F_6	Total
1990	17.9	3.5	21.5
2005	2.9	0.6	3.4
2009	1.5	0.4	1.9
2010	1.4	0.5	1.9
2011	2.7	0.8	3.5
2012	2.3	0.7	2.9
2013	2.3	0.7	3.0

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
Note: Totals may not sum due to independent rounding.

Table 4-78: PFC Emissions from Aluminum Production (kt)

Year	CF_4	C_2F_6
1990	2.4	0.3
2005	0.4	+
2009	0.2	+
2010	0.2	+
2011	0.4	0.1
2012	0.3	0.1
2013	0.3	0.1

+ Does not exceed 0.05 kt.

In 2013, U.S. primary aluminum production totaled approximately 1.9 million metric tons, a 6 percent decrease from 2012 production levels (USAA 2014). In 2013, five companies managed production at ten operational primary aluminum smelters. Three smelters were closed temporarily for the entire year in 2013 (USGS 2014). During 2013, monthly U.S. primary aluminum production was lower for every month in 2013, when compared to the corresponding months in 2012 (USAA 2014).

For 2014, total production was approximately 1.7 million metric tons compared to 1.9 million metric tons in 2013, a 12 percent decrease (USAA 2014). Based on the decrease in production, process CO_2 and PFC emissions are likely to be lower in 2014 compared to 2013 if there are no significant changes in process controls at operational facilities.

Methodology

Process CO_2 and perfluorocarbon (PFC)—i.e., perfluoromethane (CF_4) and perfluoroethane (C_2F_6)—emission estimates from primary aluminum production for 2010 through 2013 are available from EPA’s GHGRP—Subpart F (Aluminum Production) (EPA 2014). Under EPA’s GHGRP, facilities began reporting primary aluminum production process emissions (for 2010) in 2011; as a result, GHGRP data (for 2010 through 2013) are available to be incorporated into the Inventory. EPA’s GHGRP mandates that all facilities that contain an aluminum production process must report: CF_4 and C_2F_6 emissions from anode effects in all prebake and Søderberg electrolysis cells, carbon dioxide (CO_2) emissions from anode consumption during electrolysis in all prebake and Søderberg cells, and all CO_2 emissions from onsite anode baking. To estimate the process emissions, EPA’s GHGRP uses the process-specific equations (and certain technology-specific defaults) detailed in subpart F (aluminum production).[179] These equations are based on the Tier 2/Tier 3 IPCC (2006) methods for primary aluminum production, and Tier 1 methods when estimating missing data elements. It should be noted that the same methods (i.e., *2006 IPCC Guidelines*) were used for estimating the emissions prior to the availability of the reported GHGRP data in the Inventory.

Process CO_2 Emissions from Anode Consumption and Anode Baking

CO_2 emission estimates for the years prior to the introduction of EPA's GHGRP in 2010 were estimated with IPCC (2006) methods, but individual facility reported data were combined with process-specific emissions modeling. These estimates were based on information previously gathered from EPA's Voluntary Aluminum Industrial Partnership (VAIP) program, U.S. Geological Survey (USGS) Mineral Commodity reviews, and The Aluminum Association (USAA) statistics, among other sources. Since pre- and post-GHGRP estimates use the same methodology, emission estimates are comparable across the time series.

Most of the CO_2 emissions released during aluminum production occur during the electrolysis reaction of the C anode, as described by the following reaction:

$$2Al_2O_3 + 3C \rightarrow 4Al + 3CO_2$$

For prebake smelter technologies, CO_2 is also emitted during the anode baking process. These emissions can account for approximately 10 percent of total process CO_2 emissions from prebake smelters.

Depending on the availability of smelter-specific data, the CO_2 emitted from electrolysis at each smelter was estimated from: (1) The smelter's annual anode consumption, (2) the smelter's annual aluminum production and rate of anode consumption (per ton of aluminum produced) for previous and/or following years, or, (3) the smelter's annual aluminum production and IPCC default CO_2 emission factors. The first approach tracks the consumption and carbon content of the anode, assuming that all C in the anode is converted to CO_2. Sulfur, ash, and other impurities in the anode are subtracted from the anode consumption to arrive at a C consumption figure. This approach corresponds to either the IPCC Tier 2 or Tier 3 method, depending on whether smelter-specific data on anode impurities are used. The second approach interpolates smelter-specific anode consumption rates to estimate emissions during years for which anode consumption data are not available. This approach avoids substantial errors and discontinuities that could be introduced by reverting to Tier 1 methods for those years. The last approach corresponds to the IPCC Tier 1 method (2006), and is used in the absence of present or historic anode consumption data.

The equations used to estimate CO_2 emissions in the Tier 2 and 3 methods vary depending on smelter type (IPCC 2006). For Prebake cells, the process formula accounts for various parameters, including net anode consumption, and the sulfur, ash, and impurity content of the baked anode. For anode baking emissions, the formula accounts for packing coke consumption, the sulfur and ash content of the packing coke, as well as the pitch content and weight of baked anodes produced. For Søderberg cells, the process formula accounts for the weight of paste consumed per metric ton of aluminum produced, and pitch properties, including sulfur, hydrogen, and ash content.

Through the VAIP, anode consumption (and some anode impurity) data have been reported for 1990, 2000, 2003, 2004, 2005, 2006, 2007, 2008, and 2009. Where available, smelter-specific process data reported under the VAIP were used; however, if the data were incomplete or unavailable, information was supplemented using industry average values recommended by IPCC (2006). Smelter-specific CO_2 process data were provided by 18 of the 23 operating smelters in 1990 and 2000, by 14 out of 16 operating smelters in 2003 and 2004, 14 out of 15 operating smelters in 2005, 13 out of 14 operating smelters in 2006, 5 out of 14 operating smelters in 2007 and 2008, and 3 out of 13 operating smelters in 2009. For years where CO_2 emissions data or CO_2 process data were not reported by these companies, estimates were developed through linear interpolation, and/or assuming representative (e.g., previously reported or industry default) values.

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[179] *See* Code of Federal Regulations, Title 40: Protection of Environment, Part 98: Mandatory Greenhouse Gas Reporting, Subpart F—Aluminum Production. Available online at: <www.epa.gov/ghgreporting/documents/pdf/infosheets/aluminumproduction.pdf>.

In the absence of any previous historical smelter specific process data (i.e., 1 out of 13 smelters in 2009, 1 out of 14 smelters in 2006, 2007, and 2008, 1 out of 15 smelters in 2005, and 5 out of 23 smelters between 1990 and 2003), CO_2 emission estimates were estimated using Tier 1 Søderberg and/or Prebake emission factors (metric ton of CO_2 per metric ton of aluminum produced) from IPCC (2006).

Process PFC Emissions from Anode Effects

Smelter-specific PFC emissions from aluminum production for 2010 through 2013 were reported to EPA under its GHGRP. To estimate their PFC emissions and report them under EPA's GHGRP, smelters use an approach identical to the Tier 3 approach in the *2006 IPCC Guidelines* (IPCC 2006). Specifically, they use a smelter-specific slope coefficient as well as smelter-specific operating data to estimate an emission factor using the following equation:

$$\text{PFC } (CF_4 \text{ or } C_2F_6) \text{ kg/metric ton Al} = S \times (\text{Anode Effect Minutes/Cell-Day})$$

where,

S = Slope coefficient ((kg PFC/metric ton Al)/(Anode Effect Minutes/Cell-Day))
Anode Effect Minutes/Cell-Day = (Anode Effect Frequency/Cell-Day) × Anode Effect Duration (minutes)

They then multiply this emission factor by aluminum production to estimate PFC emissions. All U.S. aluminum smelters are required to report their emissions under EPA's GHGRP.

PFC emissions for the years prior to 2010 were estimated using the same equation, but the slope-factor used for some smelters was technology-specific rather than smelter-specific, making the method a Tier 2 rather than a Tier 3 approach for those smelters. Emissions and background data were reported to EPA under the VAIP. For 1990 through 2009, smelter-specific slope coefficients were available and were used for smelters representing between 30 and 94 percent of U.S. primary aluminum production. The percentage changed from year to year as some smelters closed or changed hands and as the production at remaining smelters fluctuated. For smelters that did not report smelter-specific slope coefficients, IPCC technology-specific slope coefficients were applied (IPCC 2006). The slope coefficients were combined with smelter-specific anode effect data collected by aluminum companies and reported under the VAIP to estimate emission factors over time. For 1990 through 2009, smelter-specific anode effect data were available for smelters representing between 80 and 100 percent of U.S. primary aluminum production. Where smelter-specific anode effect data were not available, representative values (e.g., previously reported or industry averages) were used.

For all smelters, emission factors were multiplied by annual production to estimate annual emissions at the smelter level. For 1990 through 2009, smelter-specific production data were available for smelters representing between 30 and 100 percent of U.S. primary aluminum production. (For the years after 2000, this percentage was near the high end of the range.) Production at non-reporting smelters was estimated by calculating the difference between the production reported under VAIP and the total U.S. production supplied by USGS or USAA, and then allocating this difference to non-reporting smelters in proportion to their production capacity. Emissions were then aggregated across smelters to estimate national emissions.

Between 1990 and 2009, production data were provided under the VAIP by 21 of the 23 U.S. smelters that operated during at least part of that period. For the non-reporting smelters, production was estimated based on the difference between reporting smelters and national aluminum production levels (from USGS and USAA), with allocation to specific smelters based on reported production capacities (from USGS).

National primary aluminum production data for 2013 were obtained via The Aluminum Association (USAA 2014). For 1990 through 2001, and 2006 (see Table 4-79) data were obtained from USGS Mineral Industry Surveys: Aluminum Annual Report (USGS 1995, 1998, 2000, 2001, 2002, 2007). For 2002 through 2005, and 2007 through 2011, national aluminum production data were obtained from the USAA's Primary Aluminum Statistics (USAA 2004–2006, 2008–2013).

Table 4-79: Production of Primary Aluminum (kt)

Year	kt
1990	4,048
2005	2,478
2009	1,727
2010	1,727
2011	1,986
2012	2,070
2013	1,948

Uncertainty and Time Series Consistency

Uncertainty was assigned to the CO_2, CF_4, and C_2F_6 emission values reported by each individual facility to EPA's GHGRP. As previously mentioned, the methods for estimating emissions for EPA's GHGRP and this report are the same, and follow the IPCC (2006) methodology. As a result, it was possible to assign uncertainty bounds (and distributions) based on an analysis of the uncertainty associated with the facility-specific emissions estimated for previous Inventory years. Uncertainty surrounding the reported CO_2, CF_4, and C_2F_6 emission values were determined to have a normal distribution with uncertainty ranges of ±6, ±16, and ±20 percent, respectively. A Monte Carlo analysis was applied to estimate the overall uncertainty of the CO_2, CF_4, and C_2F_6 emission estimates for the U.S. aluminum industry as a whole, and the results are provided below.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-80. Aluminum production-related CO_2 emissions were estimated to be between 3.2 and 3.3 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 2 percent below to 2 percent above the emission estimate of 3.3 MMT CO_2 Eq. Also, production-related CF_4 emissions were estimated to be between 2.2 and 2.4 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 6 percent below to 7 percent above the emission estimate of 2.3 MMT CO_2 Eq. Finally, aluminum production-related C_2F_6 emissions were estimated to be between 0.6 and 0.7 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 11 percent below to 11 percent above the emission estimate of 0.7 MMT CO_2 Eq.

Table 4-80: Approach 2 Quantitative Uncertainty Estimates for CO_2 and PFC Emissions from Aluminum Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Aluminum Production	CO_2	3.3	3.2	3.3	−2%	+2%
Aluminum Production	CF_4	2.3	2.2	2.4	−6%	+7%
Aluminum Production	C_2F_6	0.7	0.6	0.7	−11%	+11%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

QA/QC and Verification

Tier 1 and Tier 2 QA/QC activities were conducted consistent with the U.S. QA/QC plan. Source-specific quality control measures for Aluminum Production included checking input data, documentation, and calculations to ensure data were properly handled through the inventory process. Errors that were found during this process were corrected as necessary.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous Inventory reports) which results in time-series recalculations for most Inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2-equivalent emissions from PFCs. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

As a result, emission estimates for each year from 1990 to 2012 increased by 14 percent for CF_4, and increased by 33 percent for C_2F_6, relative to the emission estimates in the previous Inventory report.

Planned Improvements

Future improvements involve plans to replace proxy (e.g., interpolated) data with additional historical VAIP data that recently became available in order to calculate more accurate PFC emission estimates for the historical time series.

4.19 Magnesium Production and Processing (IPCC Source Category 2C4)

The magnesium metal production and casting industry uses sulfur hexafluoride (SF_6) as a cover gas to prevent the rapid oxidation of molten magnesium in the presence of air. Sulfur hexafluoride has been used in this application around the world for more than thirty years. A dilute gaseous mixture of SF_6 with dry air and/or CO_2 is blown over molten magnesium metal to induce and stabilize the formation of a protective crust. A small portion of the SF_6 reacts with the magnesium to form a thin molecular film of mostly magnesium oxide and magnesium fluoride. The amount of SF_6 reacting in magnesium production and processing is considered to be negligible and thus all SF_6 used is assumed to be emitted into the atmosphere. , Alternative cover gases, such as AM-cover™ (containing HFC-134a), Novec™ 612 (FK-5-1-12) and dilute SO_2 systems can, and are being used by some facilities in the United States. However, many facilities in the United States are still using traditional SF_6 cover gas systems.

The magnesium industry emitted 1.4 MMT CO_2 Eq. (0.06 kt) of SF_6, 0.08 MMT CO_2 Eq. (0.06 kt) of HFC-134a, and 0.002 MMT CO_2 Eq. (2.1 kt) of CO_2, in 2013. This represents a decrease of approximately 8 percent from total 2012 emissions (see Table 4-81). The decrease can be attributed to reduction in primary, secondary, and die casting SF_6 emissions between 2012 and 2013 as reported through EPA's GHGRP, with the largest absolute reduction being seen for secondary emissions. The reduction in SF_6 emissions is likely due in part to decreased production from reporting facilities in 2013. The decrease in SF_6 emissions can also be attributed by continuing industry efforts to utilize SF_6 alternatives, such as HFC-134a, Novec™612 and SO_2, to reduce greenhouse gas emissions. In 2013, total HFC-134a emissions increased from 0.01 MMT CO_2 Eq. to 0.08 MMT CO_2 Eq., while the FK 5-1-12 emissions were constant. The emissions of carrier gas, CO_2, also decreased from 2.3 kt in 2012 to 2.1 kt in 2013.

Table 4-81: SF_6, HFC-134a, FK 5-1-12 and CO_2 Emissions from Magnesium Production and Processing (MMT CO_2 Eq.)

Year	1990	2005	2009	2010	2011	2012	2013
SF_6	5.2	2.7	1.6	2.1	2.8	1.6	1.4
HFC-134a	0.0	0.0	+	+	+	+	0.1
CO_2	+	+	+	+	+	+	+
FK 5-1-12	*0.0*	*0.0*	+	+	+	+	+
Total[a]	**5.2**	**2.8**	**1.7**	**2.1**	**2.8**	**1.7**	**1.5**

Note: Emission values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
Note: Totals may not sum due to independent rounding.
+ Does not exceed 0.05 MMT CO_2 Eq.
[a] Total does not include FK 5-1-12. Values shown for informational purposes only.

Table 4-82: SF_6, HFC-134a, FK 5-1-12 and CO_2 Emissions from Magnesium Production and Processing (kt)

Year	1990		2005		2009	2010	2011	2012	2013
SF_6	0.2		0.1		0.1	0.1	0.1	0.1	0.1
HFC-134a	0.0		0.0		+	+	+	+	0.1
CO_2	1.4		2.9		1.2	1.3	3.1	2.3	2.1
FK 5-1-12	*0.0*		*0.0*		+	+	+	+	+

+ Does not exceed 0.5 kt

Methodology

Emission estimates for the magnesium industry incorporate information provided by some industry participants in EPA's SF_6 Emission Reduction Partnership for the Magnesium Industry as well as emissions data reported through subpart T (Magnesium Production and Processing) of the EPA's GHGRP. The Partnership started in 1999 and, in 2010, participating companies represented 100 percent of U.S. primary and secondary production and 16 percent of the casting sector production (i.e., die, sand, permanent mold, wrought, and anode casting). SF_6 Emissions for 1999 through 2010 from primary production, secondary production (i.e., recycling), and die casting were generally reported by Partnership participants. Partners reported their SF_6 consumption, which was assumed to be equivalent to emissions. Along with SF_6, some Partners also reported their HFC-134a and FK 5-1-12 usage, which is assumed to be equal to emissions. 2010 was the last reporting year under the Partnership. Emissions data for 2011 through 2013 were obtained through EPA's GHGRP. Under the program, owners or operators of facilities that have a magnesium production or casting process must report emissions from use of cover or carrier gases, which include SF_6, HFC-134a, FK 5-1-12 and CO_2. Consequently, cover and carrier gas emissions from magnesium production and processing were estimated for three time periods, depending on the source of the emissions data: 1990 through 1998, 1999 through 2010, and 2011 through 2013. The methodologies described below also make use of magnesium production data published by the U.S. Geological Survey (USGS).

1990 through 1998

To estimate emissions for 1990 through 1998, industry SF_6 emission factors were multiplied by the corresponding metal production and consumption (casting) statistics from USGS. For this period, it was assumed that there was no use of HFC-134a or FK 5-1-12 cover gases and hence emissions were not estimated for these alternatives.

SF_6 emission factors from 1990 through 1998 were based on a number of sources and assumptions. Emission factors for primary production were available from U.S. primary producers for 1994 and 1995. The primary production emission factors were 1.2 kg SF_6 per metric ton for 1990 through 1993, and 1.1 kg SF_6 per metric ton for 1994 through 1997. The emission factor for secondary production from 1990 through 1998 was assumed to be constant at the 1999 average Partner value. Emission factor for die casting of 4.1 kg SF_6 per metric ton was available for the mid-1990s from an international survey (Gjestland & Magers 1996) that was used for years 1990 through 1996. For 1996 through 1998, the emission factor for die casting was assumed to decline linearly to the level estimated based on Partner reports in 1999. This assumption is consistent with the trend in SF_6 sales to the magnesium sector that is reported in the RAND survey of major SF_6 manufacturers, which shows a decline of 70 percent from 1996 to 1999 (RAND 2002). Sand casting emission factors for 1990 through 2001 were assumed to be the same as the 2002 emission factor. The emission factors for the other processes (i.e., permanent mold, wrought, and anode casting), about which less is known, were assumed to remain constant at levels defined in Table 4-81. These emission factors for the other processes (i.e., permanent mold, wrought, and anode casting) were based on discussions with industry representatives.

The quantities of CO_2 carrier gas used for each production type have been estimated using the 1999 estimated CO_2 emissions data and the annual calculated rate of change of SF_6 use in the 1990 through 1999 time period. For each year and production type, the rate of change of SF_6 use between the current year and the subsequent year was first estimated. This rate of change is then applied to the CO_2 emissions of the subsequent year to determine the CO_2 emission of the current year. The emissions of carrier gases for permanent mold, wrought and anode processes are not estimated in this Inventory.

1999 through 2010

The 1999 through 2010 emissions from primary and secondary production are based on information provided by EPA's industry Partners. In some instances, there were years of missing Partner data, including SF_6 consumption and metal processed. For these situations, emissions were estimated through interpolation where possible, or by holding company-reported emissions (as well as production) constant from the previous year. For alternative cover gases, including HFC-134a and FK 5-1-12, mainly reported data was relied upon. That is, unless a Partner reported using an alternative cover gas, it was not assumed it was used. Emissions of alternate gases were also estimated through linear interpolation where possible.

The die casting emission estimates for 1999 through 2010 are also based on information supplied by industry Partners. When a Partner was determined to be no longer in production, its metal production and usage rates were set to zero. Missing data on emissions or metal input was either interpolated or held constant at the last available reported value. In 1999 and from 2008 through 2010, Partners did not account for all die casting tracked by USGS, and, therefore, it was necessary to estimate the emissions of die casters who were not Partners. For 1999, die casters who were not Partners were assumed to be similar to Partners who cast small parts. Due to process requirements, these casters consume larger quantities of SF_6 per metric ton of processed magnesium than casters that process large parts. Consequently, emission estimates from this group of die casters were developed using an average emission factor of 5.2 kg SF_6 per metric ton of magnesium. This emission factor was developed using magnesium production and SF_6 usage data for the year 1999. For 2008 through 2010, the characteristics of the die casters who were not Partners were not well known, and therefore the emission factor for these die casters was set equal to 3.0 kg SF_6 per metric ton of magnesium, the average of the emission factors reported over the same period by the die casters who were Partners.

The emissions from other casting operations were estimated by multiplying emission factors (kg SF_6 per metric ton of metal produced or processed) by the amount of metal produced or consumed from USGS, with the exception of some years for which Partner sand casting emissions data are available. The emission factors for sand casting activities were acquired through the data reported by the Partnership for 2002 to 2006. For 1999-2001, the sand casting emission factor was held constant at the 2002 Partner-reported level. For 2007 through 2010, the sand casting Partner did not report and the reported emission factor from 2005 was applied to the Partner and to all other sand casters.

The emission factors for primary production, secondary production and sand casting for the 1999 to 2010 are not published to protect company-specific production information. However, the emission factor for primary production has not risen above the average 1995 Partner value of 1.1 kg SF_6 per metric ton. The emission factors for the other industry sectors (i.e., permanent mold, wrought, and anode casting) were based on discussions with industry representatives. The emission factors for casting activities are provided below in Table 4-83.

The emissions of HFC-134a and FK-5-1-12 were included in the estimates for only instances where Partners reported that information to the Partnership. Emissions of these alternative cover gases were not estimated for instances where emissions were not reported.

CO_2 carrier gas emissions were estimated using the emission factors developed based on GHGRP-reported carrier gas and cover gas data, by production type. It was assumed that the use of carrier gas, by production type, is proportional to the use of cover gases. Therefore, an emission factor, in kg CO_2 per kg cover gas and weighted by the cover gases used, was developed for each of the production types. GHGRP data on which these emissions factors are based was available for primary, secondary, die casting and sand casting. The emission factors were applied to the total quantity of all cover gases used (SF_6, HFC-134a, and FK5-1-12) by production type in this time period. Carrier gas emissions for the 1999 through 2010 time period were only estimated for those Partner companies that reported using CO_2 as a carrier gas through the GHGRP. Using this approach helped ensure time series consistency. The emissions of carrier gases for permanent mold, wrought and anode processes are not estimated in this Inventory.

Table 4-83: SF_6 Emission Factors (kg SF_6 per metric ton of magnesium)

Year	Die Casting[a]	Permanent Mold	Wrought	Anodes
1999	2.14[b]	2	1	1
2000	0.72	2	1	1
2001	0.72	2	1	1
2002	0.71	2	1	1
2003	0.81	2	1	1
2004	0.79	2	1	1
2005	0.77	2	1	1
2006	0.88	2	1	1
2007	0.64	2	1	1
2008	0.10	2	1	1
2009	2.30	2	1	1
2010	2.94	2	1	1

[a] Weighted average includes all die casters, Partners and non-Partners. For the majority of the time series (2000-2007), Partners made up 100 percent of die casters in the U.S.

[b] Weighted average that includes an estimated emission factor of 5.2 kg SF_6 per metric ton of magnesium for die casters that do not participate in the Partnership.

2011 through 2013

For 2011 through 2013, for the primary and secondary producers, GHGRP-reported cover and carrier gases emissions data were used. For die and sand casting, some emissions data was obtained through EPA's GHGRP. The balance of the emissions for these industry segments were estimated based on previous Partner reporting (i.e., for Partners that did not report emissions through EPA's GHGRP) or were estimated by multiplying emission factors by the amount of metal produced or consumed. Partners who did not report through EPA's GHGRP were assumed to have continued to emit SF_6 at the last reported level, which was from 2010 in most cases. All Partners were assumed to have continued to consume magnesium at the last reported level. Where the total metal consumption estimated for the Partners fell below the U.S. total reported by USGS, the difference was multiplied by the emission factors discussed in the section above. For the other types of production and processing (i.e., permanent mold, wrought, and anode casting), emissions were estimated by multiplying the industry emission factors with the metal production or consumption statistics obtained from USGS. For 2013, pre-published USGS consumption statistics were obtained via communications with USGS (USGS 2013)

Uncertainty and Time Series Consistency

Uncertainty surrounding the total estimated emissions in 2013 is attributed to the uncertainties around SF_6, HFC-134a and CO_2 emission estimates. To estimate the uncertainty surrounding the estimated 2013 SF_6 emissions from magnesium production and processing, the uncertainties associated with three variables were estimated: (1) emissions reported by magnesium producers and processors for 2013 through EPA's GHGRP, (2) emissions estimated for magnesium producers and processors that reported via the Partnership in prior years but did not report 2013 emissions through EPA's GHGRP, and (3) emissions estimated for magnesium producers and processors that did not participate in the Partnership or report through EPA's GHGRP. An uncertainty of 5 percent was assigned to the emissions (usage) data reported by each GHGRP reporter for all the cover and carrier gases (per the *2006 IPCC Guidelines*). If facilities did not report emissions data during the current reporting year through EPA's GHGRP reporting program, SF_6 emissions data were held constant at the most recent available value reported through the Partnership. The uncertainty associated with these values was estimated to be 30 percent for each year of extrapolation. Alternate cover gas and carrier gases data was set equal to zero if the facilities did not report via the GHGRP program. One known sand caster (the lone Partner) has not reported since 2007 and its activity and emission factor were held constant at 2005 levels due to a reporting anomaly in 2006 because of malfunctions at the facility. The uncertainty associated with the SF_6 usage for the sand casting Partner was 85 percent. For those industry processes that are not represented in the Partnership, such as permanent mold and wrought casting, SF_6 emissions were estimated using production and consumption statistics reported by USGS and estimated process-specific emission factors (see Table 4-84). The uncertainties associated with the emission factors and USGS-reported statistics were assumed to be 75 percent and 25 percent, respectively. Emissions associated with die casting and sand casting activities utilized emission factors based on Partner reported data with an uncertainties of 75 percent. In general, where precise quantitative information was not available on the uncertainty of a parameter, a conservative (upper-bound) value was used.

Additional uncertainties exist in these estimates that are not addressed in this methodology, such as the basic assumption that SF_6 neither reacts nor decomposes during use. The melt surface reactions and high temperatures associated with molten magnesium could potentially cause some gas degradation. Previous measurement studies have identified SF_6 cover gas degradation in die casting applications on the order of 20 percent (Bartos et al. 2007). Sulfur hexafluoride may also be used as a cover gas for the casting of molten aluminum with high magnesium content; however, the extent to which this technique is used in the United States is unknown.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-84. Total emissions associated with magnesium production and processing were estimated to be between 1.3 and 1.7 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 11 percent below to 12 percent above the 2013 emission estimate of 1.5 MMT CO_2 Eq. The uncertainty estimates for 2013 are similar relative to the uncertainty reported for 2012 in the previous Inventory report.

Table 4-84: Approach 2 Quantitative Uncertainty Estimates for SF_6, HFC-134a and CO_2 Emissions from Magnesium Production and Processing (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Magnesium Production	SF_6, HFC-134a, CO_2	1.5	1.3	1.7	-11%	+12%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

In the current Inventory, emission estimates for alternate cover gases and carrier gas has been incorporated as the information is now available from EPA's GHGRP. The alternative cover gases have lower GWPs than SF_6, and tend to quickly degrade during their exposure to the molten metal. Magnesium producers and processors began using these cover gases starting in around 2006, as based on Partnership reported data. The amounts being used by companies on the whole are low and have a minor effect on the overall emissions from the industry. This is also attributable to their relatively lower GWPs. SF_6 has a GWP of 22,800, whereas HFC-134a has a GWP of 1,430. Similarly, EPA's GHGRP now provides access to data on carrier gases, allowing for this information to be integrated in the Inventory. Emissions of CO_2 have also been included in the total emissions from the industry. This has led to slight increase in overall emissions for each year compared to the previous Inventory. CO_2 carrier gas emissions have been included across the entire time series to ensure time series consistency.

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2-equivalent emissions from CH_4, HFCs, and PFCs. The GWPs of N_2O and SF_6 have decreased, leading to a decrease in CO_2-equivalent emissions for SF_6. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

As a net result, emission estimates for each year from 1990 to 2013 have slightly decreased, relative to the previous Inventory report.

For one facility, a recalculation for 2011 SF_6 emissions was performed to ensure methodological consistency. The emissions for this facility and year were previously estimated using a company-specific growth rate based on data reported through the Partnership. This estimate has been revised by interpolating the reported emissions between 2010 and 2012, reported via the Partnership and EPA's GHGRP respectively. This has caused a slight increase in the SF_6 emissions for 2011 compared to the previous Inventory.

Planned Improvements

Cover gas research conducted over the last decade has found that SF_6 used for magnesium melt protection can have degradation rates on the order of 20 percent in die casting applications (Bartos et al. 2007). Current emission estimates assume (per the *2006 IPCC Guidelines*) that all SF_6 utilized is emitted to the atmosphere. Additional research may lead to a revision of the *2006 IPCC Guidelines* to reflect this phenomenon and until such time, developments in this sector will be monitored for possible application to the inventory methodology.

Usage and emission details of carrier gases in permanent mold, wrought and anode processes will be researched as part of a future inventory. Based on this research, it will be determined if CO_2 carrier gas emissions are to be estimated.

4.20 Lead Production (IPCC Source Category 2C5)

Lead production in the United States consists of both primary and secondary processes—both of which emit CO_2 (Sjardin 2003). Emissions from fuels consumed for energy purposes during the production of lead are accounted for in the Energy chapter.

Primary production of lead through the direct smelting of lead concentrate produces CO_2 emissions as the lead concentrates are reduced in a furnace using metallurgical coke (Sjardin 2003). Primary lead production, in the form of direct smelting, occurs at a just a single smelter in Missouri. This primary lead smelter was closed at the end of 2013 (USGS 2014b).

Similar to primary lead production, CO_2 emissions from secondary production result when a reducing agent, usually metallurgical coke, is added to the smelter to aid in the reduction process. Carbon dioxide emissions from secondary production also occur through the treatment of secondary raw materials (Sjardin 2003). Secondary production primarily involves the recycling of lead acid batteries and post-consumer scrap at secondary smelters. Of all the domestic secondary smelters operational in 2013, 12 smelters had capacities of 30,000 tons or more and were collectively responsible for more than 95 percent of secondary lead production in 2013 (USGS 2014a). Secondary lead production has increased in the United States over the past decade while primary lead production has decreased. In 2013, secondary lead production accounted for nearly 90 percent of total lead production. Similarly, secondary lead accounted for approximately 68 percent of total domestic lead consumption (USGS 2014a).

In 2013, total secondary lead production in the United States was slightly less than that in 2012. Domestic secondary lead producers expanded capacity and others closed plants, but total production capacity remained essentially unchanged. In April 2013, a leading producer closed its 70,000 ton capacity smelter in Reading, PA, and in September reduced production at its 90,000 ton capacity smelter in Vernon, CA, by 15 percent. Increases in exports of spent lead-acid batteries in recent years have decreased the amount of scrap available to secondary smelters (USGS 2014a).

U.S. primary lead production increased by approximately 6 percent from 2012 to 2013, and has decreased by 71 percent since 1990. In 2013, U.S. secondary lead production slightly decreased from 2012 levels by approximately 1 percent, but has increased by 19 percent since 1990 (USGS 1995 through 2013, USGS 2014a).

In 2013, U.S. primary and secondary lead production totaled 1,218,000 metric tons (USGS 2014a). The resulting emissions of CO_2 from 2013 lead production were estimated to be 0.5 MMT CO_2 Eq. (525 kt) (see Table 4-85). The majority of 2013 lead production is from secondary processes, which accounted for 94 percent of total 2013 CO_2 emissions from lead production. At last reporting, the United States was the third largest mine producer of lead in the world, behind China and Australia, accounting for approximately 6 percent of world production in 2013 (USGS 2014a).

Table 4-85: CO_2 Emissions from Lead Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	0.5	516
2005	0.6	553
2009	0.5	525
2010	0.5	542
2011	0.5	538
2012	0.5	527
2013	0.5	525

After a steady increase in total emissions from 1995 to 2000, total emissions have gradually decreased since 2000 but were still 2 percent greater in 2013 than in 1990. Although primary production has decreased significantly (71 percent since 1990), secondary production has increased by about 19 percent over the same time period. Since secondary production is more emissions-intensive, the increase in secondary production since 1990 has resulted in a net increase in emissions despite the sharp decrease in primary production (USGS 1995 through 2013; USGS 2014a).

Methodology

The methods used to estimate emissions for lead production are based on Sjardin's work (Sjardin 2003) for lead production emissions and Tier 1 methods from the *2006 IPCC Guidelines* (IPCC 2006). The Tier 1 equation is as follows:

$$CO_2\ Emissions = (DS \times EFa) + (S \times EFb)$$

Where,

DS = Lead produced by direct smelting, metric ton
S = Lead produced from secondary materials
EFa, b = Applicable emission factor, metric tons CO_2/metric ton product

For primary lead production using direct smelting, Sjardin (2003) and the IPCC (2006) provide an emission factor of 0.25 metric tons CO_2/metric ton lead. For secondary lead production, Sjardin (2003) and IPCC (2006) provide an emission factor of 0.25 metric tons CO_2/metric ton lead for direct smelting, as well as an emission factor of 0.2 metric tons CO_2/metric ton lead produced for the treatment of secondary raw materials (i.e., pretreatment of lead acid batteries). Since the secondary production of lead involves both the use of the direct smelting process and the treatment of secondary raw materials, Sjardin recommends an additive emission factor to be used in conjunction with the secondary lead production quantity. The direct smelting factor (0.25) and the sum of the direct smelting and pretreatment emission factors (0.45) are multiplied by total U.S. primary and secondary lead production, respectively, to estimate CO_2 emissions.

The 1990 through 2013 activity data for primary and secondary lead production (see Table 4-86) were obtained from the USGS (USGS 1995 through 2013; 2014a).

Table 4-86: Lead Production (Metric Tons)

Year	Primary	Secondary
1990	404,000	922,000

2005	143,000	1,150,000
2009	103,000	1,110,000
2010	115,000	1,140,000
2011	118,000	1,130,000
2012	111,000	1,110,000
2013	118,000	1,100,000

Uncertainty and Time-Series Consistency

Uncertainty associated with lead production relates to the emission factors and activity data used. The direct smelting emission factor used in primary production is taken from Sjardin (2003) who averaged the values provided by three other studies (Dutrizac et al. 2000, Morris et al. 1983, Ullman 1997). For secondary production, Sjardin (2003) added a CO_2 emission factor associated with battery treatment. The applicability of these emission factors to plants in the United States is uncertain. There is also a smaller level of uncertainty associated with the accuracy of primary and secondary production data provided by the USGS.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-87. Lead production CO_2 emissions were estimated to be between 0.4 and 0.6 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 14 percent below and 15 percent above the emission estimate of 0.5 MMT CO_2 Eq.

Table 4-87: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Lead Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Lead Production	CO_2	0.5	0.4	0.6	-14%	+15%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP to improve the emission estimates for the Lead Production source category. Particular attention would be made to ensure time series consistency of the emission estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[180]

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[180] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdffiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

4.21 Zinc Production (IPCC Source Category 2C6)

Zinc production in the United States consists of both primary and secondary processes. Of the primary and secondary processes used in the United States, only the electrothermic and Waelz kiln secondary processes result in non-energy CO_2 emissions (Viklund-White 2000). Emissions from fuels consumed for energy purposes during the production of zinc are accounted for in the Energy chapter.

The majority of zinc produced in the United States is used for galvanizing. Galvanizing is a process where zinc coating is applied to steel in order to prevent corrosion. Zinc is used extensively for galvanizing operations in the automotive and construction industry. Zinc is also used in the production of zinc alloys and brass and bronze alloys (e.g., brass mills, copper foundries, copper ingot manufacturing, etc.). Zinc compounds and dust are also used, to a lesser extent, by the agriculture, chemicals, paint, and rubber industries.

Primary production in the United States is conducted through the electrolytic process, while secondary techniques include the electrothermic and Waelz kiln processes, as well as a range of other metallurgical, hydrometallurgical, and pyrometallurgical processes. Worldwide primary zinc production also employs a pyrometallurgical process using the Imperial Smelting Furnace process; however, this process is not used in the United States (Sjardin 2003).

In the electrothermic process, roasted zinc concentrate and secondary zinc products enter a sinter feed where they are burned to remove impurities before entering an electric retort furnace. Metallurgical coke is added to the electric retort furnace as a carbon-containing reductant. This concentration step, using metallurgical coke and high temperatures, reduces the zinc oxides and produces vaporized zinc, which is then captured in a vacuum condenser. This reduction process also generates non-energy CO_2 emissions.

$$ZnO + C \rightarrow Zn(gas) + CO_2 \qquad \text{(Reaction 1)}$$

$$ZnO + CO \rightarrow Zn(gas) + CO_2 \qquad \text{(Reaction 2)}$$

In the Waelz kiln process, electric arc furnace (EAF) dust, which is captured during the recycling of galvanized steel, enters a kiln along with a reducing agent (typically carbon-containing metallurgical coke). When kiln temperatures reach approximately 1100-1200°C, zinc fumes are produced, which are combusted with air entering the kiln. This combustion forms zinc oxide, which is collected in a baghouse or electrostatic precipitator, and is then leached to remove chloride and fluoride. The use of carbon-containing metallurgical coke in a high-temperature fuming process results in non-energy CO_2 emissions. Through this process, approximately 0.33 metric ton of zinc is produced for every metric ton of EAF dust treated (Viklund-White 2000).

The only companies in the United States that use emissive technology to produce secondary zinc products are Horsehead, PIZO, and Steel Dust Recycling. For Horsehead, EAF dust is recycled in Waelz kilns at their Beaumont, TX; Calumet, IL; Palmerton, PA; Rockwood, TN; and Barnwell, SC facilities. These Waelz kiln facilities produce intermediate zinc products (crude zinc oxide or calcine), most of which is transported to their Monaca, PA facility where the products are smelted into refined zinc using electrothermic technology. Some of Horsehead's intermediate zinc products that are not smelted at Monaca are instead exported to other countries around the world (Horsehead 2010a). PIZO and Steel Dust Recycling recycle EAF dust into intermediate zinc products using Waelz kilns, and then sell the intermediate products to companies who smelt it into refined products.

In 2013, U.S. primary and secondary refined zinc production were estimated to total 250,000 metric tons (USGS 2014b) (see Table 4-88). Domestic zinc mine production increased slightly in 2013 compared to 2012 levels, primarily owing to increase in zinc production at a zinc-lead mine in Alaska and two zinc-mining complexes in Tennessee. Zinc metal production decreased by 4 percent owing to a decline in secondary production; a zinc-recycling company closed its smelter in Pennsylvania towards the end of 2013 as it began production at its new recycling facility in North Carolina starting 2014 (USGS 2014b). Primary zinc production (primary slab zinc) increased slightly in 2013, while, secondary zinc production in 2013 decreased relative to 2012.

Emissions of CO_2 from zinc production in 2013 were estimated to be 1.4 MMT CO_2 Eq. (1,429 kt) (see Table 4-89). All 2013 CO_2 emissions resulted from secondary zinc production processes. Emissions from zinc production in the

U.S. have increased overall since 1990 due to a gradual shift from non-emissive primary production to emissive secondary production. In 2013, emissions were estimated to be 126 percent higher than they were in 1990.

Table 4-88: Zinc Production (Metric Tons)

Year	Primary	Secondary
1990	262,704	95,708
2005	191,120	156,000
2009	94,000	109,000
2010	120,000	129,000
2011	110,000	138,000
2012	114,000	147,000
2013	120,000	130,000

Table 4-89: CO_2 Emissions from Zinc Production (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	0.6	632
2005	1.0	1,030
2009	0.9	943
2010	1.2	1,182
2011	1.3	1,286
2012	1.5	1,486
2013	1.4	1,429

Methodology

The methods used to estimate non-energy CO_2 emissions from zinc production using the electrothermic primary production and Waelz kiln secondary production processes are based on Tier 1 methods from the *2006 IPCC Guidelines* (IPCC 2006). The Tier 1 emission factors provided by IPCC for Waelz kiln-based secondary production were derived from coke consumption factors and other data presented in Vikland-White (2000). These coke consumption factors as well as other inputs used to develop the Waelz kiln emission factors are shown below. IPCC does not provide an emission factor for electrothermic processes due to limited information; therefore, the Waelz kiln-specific emission factors were also applied to zinc produced from electrothermic processes.

For Waelz kiln-based production, IPCC recommends the use of emission factors based on EAF dust consumption, if possible, rather than the amount of zinc produced since the amount of reduction materials used is more directly dependent on the amount of EAF dust consumed. Since only a portion of emissive zinc production facilities consume EAF dust, the emission factor based on zinc production is applied to the non-EAF dust consuming facilities while the emission factor based on EAF dust consumption is applied to EAF dust consuming facilities.

The Waelz kiln emission factor based on the amount of zinc produced was developed based on the amount of metallurgical coke consumed for non-energy purposes per ton of zinc produced (i.e., 1.19 metric tons coke/metric ton zinc produced) (Viklund-White 2000), and the following equation:

$$EF_{Waelz\ Kiln} = \frac{1.19\ metric\ tons\ coke}{metric\ tons\ zinc} \times \frac{0.85\ metric\ tons\ C}{metric\ tons\ coke} \times \frac{3.67\ metric\ tons\ CO_2}{metric\ tons\ C} = \frac{3.70\ metric\ tons\ CO_2}{metric\ tons\ zinc}$$

The Waelz kiln emission factor based on the amount of EAF dust consumed was developed based on the amount of metallurgical coke consumed per ton of EAF dust consumed (i.e., 0.4 metric tons coke/metric ton EAF dust consumed) (Viklund-White 2000), and the following equation:

$$EF_{EAF\ Dust} = \frac{0.4\ metric\ tons\ coke}{metric\ tons\ EAF\ Dust} \times \frac{0.85\ metric\ tons\ C}{metric\ tons\ coke} \times \frac{3.67\ metric\ tons\ CO_2}{metric\ tons\ C} = \frac{1.24\ metric\ tons\ CO_2}{metric\ tons\ EAF\ Dust}$$

The total amount of EAF dust consumed by Horsehead at their Waelz kilns was available from Horsehead financial reports for years 2006 through 2013 (Horsehead 2007, 2008, 2010a, 2011, 2012, 2013, and 2014). Consumption levels for 1990 through 2005 were extrapolated using the percentage change in annual refined zinc production at secondary smelters in the United States as provided by *USGS Minerals Yearbook: Zinc* (USGS 1995 through 2006). The EAF dust consumption values for each year were then multiplied by the 1.24 metric tons CO_2/metric ton EAF dust consumed emission factor to develop CO_2 emission estimates for Horsehead's Waelz kiln facilities.

The amount of EAF dust consumed by Steel Dust Recycling (SDR) and their total production capacity were obtained from SDR's facility in Alabama for the years 2011 through 2013 (Rowland 2012 and 2014). SDR's facility in Alabama underwent expansion in 2011 to include a second unit (operational since early- to mid-2012). SDR's facility has been operational since 2008. Annual consumption data for SDR was not publicly available for the years 2008, 2009, and 2010. These data were estimated using data for Horsehead's Waelz kilns for 2008-2010 (Horsehead 2007, 2008, 2010a, 2010b, and 2011). Annual capacity utilization ratios were calculated using Horsehead's annual consumption and total capacity for the years 2008-2010. Horsehead's annual capacity utilization ratios were multiplied with SDR's total capacity to estimate SDR's consumption for each of the years, 2008 through 2010 (Steel Dust Recycling LLC 2013).

PIZO Technologies Worldwide LLC's facility in Arkansas has been operational since 2009. The amount of EAF dust consumed by PIZO's facility for 2009 through 2013 was not publicly available. EAF dust consumption for PIZO's facility for 2009 and 2010 were estimated by calculating annual capacity utilization of Horsehead's Waelz kilns and multiplying this utilization ratio by PIZO's total capacity (PIZO 2012). EAF dust consumption for PIZO's facility for 2011 through 2013 were estimated by applying the average annual capacity utilization rates for Horsehead and SDR (Grupo PROMAX) to PIZO's annual capacity (Horsehead 2012, 2013, and 2014; Rowland 2012 and 2014; PIZO 2012 and 2014). The 1.24 metric tons CO_2/metric ton EAF dust consumed emission factor was then applied to PIZO's and Steel Dust Recycling's estimated EAF dust consumption to develop CO_2 emission estimates for those Waelz kiln facilities.

Refined zinc production levels for Horsehead's Monaca, PA facility (utilizing electrothermic technology) were available from the company for years 2005 through 2013 (Horsehead 2008, 2011, 2012, 2013, and 2014). Production levels for 1990 through 2004 were extrapolated using the percentage changes in annual refined zinc production at secondary smelters in the United States as provided by USGS Minerals Yearbook: Zinc (USGS 1995 through 2005). The 3.70 metric tons CO_2/metric ton zinc emission factor was then applied to the Monaca facility's production levels to estimate CO_2 emissions for the facility. The Waelz kiln production emission factor was applied in this case rather than the EAF dust consumption emission factor since Horsehead's Monaca facility did not consume EAF dust.

Uncertainty and Time-Series Consistency

The uncertainties contained in these estimates are two-fold, relating to activity data and emission factors used.

First, there is uncertainty associated with the amount of EAF dust consumed in the United States to produce secondary zinc using emission-intensive Waelz kilns. The estimate for the total amount of EAF dust consumed in Waelz kilns is based on (1) an EAF dust consumption value reported annually by Horsehead Corporation as part of its financial reporting to the Securities and Exchange Commission (SEC), and (2) an EAF dust consumption value obtained from the Waelz kiln facility operated in Alabama by Steel Dust Recycling LLC. Since actual EAF dust consumption information is not available for PIZO's facility (2009-2010) and SDR's facility (2008-2010), the amount is estimated by multiplying the EAF dust recycling capacity of the facility (available from the company's Web site) by the capacity utilization factor for Horsehead Corporation (which is available from Horsehead's financial reports). Also, the EAF dust consumption for PIZO's facility for 2011-2013 was estimated by multiplying the average capacity utilization factor developed from Horsehead Corp. and SDR's annual capacity utilization rates by PIZO's EAF dust recycling capacity. Therefore, there is uncertainty associated with the assumption used to estimate PIZO and SDR's annual EAF dust consumption values (except SDR's EAF dust consumption for 2011-2013 which were obtained from SDR's recycling facility in Alabama).

Second, there are uncertainties associated with the emission factors used to estimate CO_2 emissions from secondary zinc production processes. The Waelz kiln emission factors are based on materials balances for metallurgical coke and EAF dust consumed as provided by Viklund-White (2000). Therefore, the accuracy of these emission factors depend upon the accuracy of these materials balances. Data limitations prevented the development of emission factors for the electrothermic process. Therefore, emission factors for the Waelz kiln process were applied to both electrothermic and Waelz kiln production processes. The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-90. Zinc production CO_2 emissions were estimated to be between 1.2 and 1.7 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 16 percent below and 18 percent above the emission estimate of 1.4 MMT CO_2 Eq.

Table 4-90: Approach 2 Quantitative Uncertainty Estimates for CO_2 Emissions from Zinc Production (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Zinc Production	CO_2	1.4	1.2	1.7	-16%	+18%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time series to ensure consistency in emissions from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

In the previous version of the Inventory (i.e., 1990-2012), EAF dust consumption data for SDR's Alabama facility were not available for 2012. Therefore, 2011 data were used as proxy for 2012. During 2013 updates to the Inventory, these data were obtained from SDR (Rowland 2014). This change caused an increase of approximately 4.5 percent in the 2012 emissions.

Planned Improvements

Future improvements involve evaluating and analyzing data reported under EPA's GHGRP to improve the emission estimates for the Zinc Production source category. Particular attention would be made to ensure time series consistency of the emissions estimates presented in future Inventory reports, consistent with IPCC and UNFCCC guidelines. This is required as the facility-level reporting data from EPA's GHGRP, with the program's initial requirements for reporting of emissions in calendar year 2010, are not available for all inventory years (i.e., 1990 through 2009) as required for this Inventory. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[181]

4.22 Semiconductor Manufacture (IPCC Source Category 2E1)

The semiconductor industry uses multiple long-lived fluorinated greenhouse gases in plasma etching and plasma enhanced chemical vapor deposition (PECVD) processes to produce semiconductor products. The gases most commonly employed are trifluoromethane (HFC-23 or CHF_3), perfluoromethane (CF_4), perfluoroethane (C_2F_6),

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[181] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdffiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

nitrogen trifluoride (NF_3), sulfur hexafluoride (SF_6), and nitrous oxide (N_2O), although other compounds such as perfluoropropane (C_3F_8) and perfluorocyclobutane (c-C_4F_8) are also used. The exact combination of compounds is specific to the process employed.

A single 300 mm silicon wafer that yields between 400 to 600 semiconductor products (devices or chips) may require more than 100 distinct fluorinated-gas-using process steps, principally to deposit and pattern dielectric films. Plasma etching (or patterning) of dielectric films, such as silicon dioxide and silicon nitride, is performed to provide pathways for conducting material to connect individual circuit components in each device. The patterning process uses plasma-generated fluorine atoms, which chemically react with exposed dielectric film to selectively remove the desired portions of the film. The material removed as well as undissociated fluorinated gases flow into waste streams and, unless emission abatement systems are employed, into the atmosphere. PECVD chambers, used for depositing dielectric films, are cleaned periodically using fluorinated and other gases. During the cleaning cycle the gas is converted to fluorine atoms in plasma, which etches away residual material from chamber walls, electrodes, and chamber hardware. Undissociated fluorinated gases and other products pass from the chamber to waste streams and, unless abatement systems are employed, into the atmosphere.

In addition to emissions of unreacted gases, some fluorinated compounds can also be transformed in the plasma processes into different fluorinated compounds which are then exhausted, unless abated, into the atmosphere. For example, when C_2F_6 is used in cleaning or etching, CF_4 is generated and emitted as a process by-product. Besides dielectric film etching and PECVD chamber cleaning, much smaller quantities of fluorinated gases are used to etch polysilicon films and refractory metal films like tungsten.

Nitrous oxide is used in manufacturing semiconductor devices to produce thin films by CVD and nitridation processes as well as for N-doping of compound semiconductors and reaction chamber conditioning (Doering 2000).

For 2013, total CO_2 weighted emissions of all fluorinated greenhouse gases and nitrous oxide by the U.S. semiconductor industry were estimated to be 4.2 MMT CO_2 Eq. Combined emissions of all greenhouse gases are presented in Table 4-91 and Table 4-92 below for years 1990, 2005 and the period 2009 to 2013. The rapid growth of this industry and the increasing complexity (growing number of layers[182]) of semiconductor products led to an increase in emissions of 153 percent between 1990 and 1999, when emissions peaked at 9.1 MMT CO_2 Eq. The emissions growth rate began to slow after 1999, and emissions declined by 54 percent between 1999 and 2013. Together, industrial growth, adoption of emissions reduction technologies, including but not limited to abatement technologies, and shift in gas usages resulted in a net increase in emissions of 16 percent between 1990 and 2013.

There was a sizable dip seen in emissions between 2008 and 2009, a 28 percent decrease, due to the slowed economic growth, and hence production, during this time. The industry recovered and emissions rose between 2009 and 2010 by more than 25 percent and between 2010 and 2011 by 29 percent; reductions in emissions were observed between 2011 and 2012, and 2012 and 2013 at 9 percent and 7 percent, respectively.

Table 4-91: PFC, HFC, SF_6, NF_3, and N_2O Emissions from Semiconductor Manufacture (MMT CO_2 Eq.)

Year	1990		2005		2009	2010	2011	2012	2013
CF_4	0.8		1.1		0.8	1.1	1.4	1.3	1.2
C_2F_6	2.0		1.9		1.1	1.4	1.8	1.6	1.5
C_3F_8	+		0.1		0.1	0.1	0.2	0.1	0.1
C_4F_8	+		0.1		+	+	0.1	0.1	0.1
HFC-23	0.2		0.2		0.2	0.2	0.2	0.2	0.2
SF_6	0.5		0.7		0.3	0.4	0.4	0.4	0.4
NF_3	+		0.5		0.4	0.5	0.7	0.6	0.6
Total F-GHGs	**3.6**		**4.6**		**2.9**	**3.7**	**4.7**	**4.3**	**4.0**
N_2O	+		0.1		0.1	0.1	0.2	0.2	0.2

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[182] Complexity is a term denoting the circuit required to connect the active circuit elements (transistors) on a chip. Increasing miniaturization, for the same chip size, leads to increasing transistor density, which, in turn, requires more complex interconnections between those transistors. This increasing complexity is manifested by increasing the levels (i.e., layers) of wiring, with each wiring layer requiring fluorinated gas usage for its manufacture.

Total	**3.6**	**4.7**	**3.1**	**3.8**	**4.9**	**4.5**	**4.2**

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
Note: Totals may not sum due to independent rounding.
+ Does not exceed 0.05 MMT CO_2 Eq.

Table 4-92: PFC, HFC, SF_6, NF_3, and N_2O Emissions from Semiconductor Manufacture (kt)

Year	1990	2005	2009	2010	2011	2012	2013
CF_4	0.11	0.14	0.11	0.14	0.19	0.17	0.16
C_2F_6	0.16	0.16	0.09	0.11	0.14	0.13	0.12
C_3F_8	+	+	+	+	+	+	+
C_4F_8	+	+	+	+	+	+	+
HFC-23	+	+	+	+	+	+	+
SF_6	+	+	+	+	+	+	+
NF_3	+	+	+	+	+	+	+
N_2O	0.12	0.41	0.45	0.49	0.79	0.65	0.61

+ Does not exceed 0.05 kt

Methodology

Emissions are based on data reported through Subpart I, Electronics Manufacture, of EPA's GHGRP, Partner reported emissions data received through the EPA's PFC[183] Reduction/Climate Partnership, EPA's PFC Emissions Vintage Model (PEVM)—a model that estimates industry emissions in the absence of emission control strategies (Burton and Beizaie 2001)[184], and estimates of industry activity (i.e., total manufactured layer area). The availability and applicability of reported data from the EPA Partnership and EPA's GHGRP differs across the 1990 through 2013 time series. Consequently, F-GHG emissions from semiconductor manufacturing were estimated using five distinct methods, one each for the periods 1990 through 1994, 1995 through 1999, 2000 through 2006, 2007 through 2010, and 2011 through 2013. N_2O emissions were estimated using three distinct methods, one each for the period 1990 through 1994, 1995 through 2010, and 2011 through 2013.

1990 through 1994

From 1990 through 1994, Partnership data were unavailable and emissions were modeled using the PEVM (Burton and Beizaie 2001).[185] The 1990 to 1994 emissions are assumed to be uncontrolled, since reduction strategies such as chemical substitution and abatement were yet to be developed.

PEVM is based on the recognition that fluorinated greenhouse gas emissions from semiconductor manufacturing vary with: (1) the number of layers that comprise different kinds of semiconductor devices, including both silicon wafer and metal interconnect layers, and (2) silicon consumption (i.e., the area of semiconductors produced) for each kind of device. The product of these two quantities, Total Manufactured Layer Area (TMLA), constitutes the activity data for semiconductor manufacturing. PEVM also incorporates an emission factor that expresses emissions per unit of layer-area. Emissions are estimated by multiplying TMLA by this emission factor.

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[183] In the context of the EPA Partnership and PEVM, PFC refers to perfluorocompounds, not perfluorocarbons.

[184] A Partner refers to a participant in the U.S. EPA PFC Reduction/Climate Partnership for the Semiconductor Industry. Through a Memorandum of Understanding (MoU) with the EPA, Partners voluntarily reported their PFC emissions to the EPA by way of a third party, which aggregated the emissions through 2010. For 2011, while no MOU existed, it was assumed that the same companies that were Partners in 2010 were "Partners" in 2011 for purposes of estimating inventory emissions.

[185] Various versions of the PEVM exist to reflect changing industrial practices. From 1990 to 1994 emissions estimates are from PEVM v1.0, completed in September 1998. The emission factor used to estimate 1990 to 1994 emissions is an average of the 1995 and 1996 emissions factors, which were derived from Partner reported data for those years.

PEVM incorporates information on the two attributes of semiconductor devices that affect the number of layers: (1) linewidth technology (the smallest manufactured feature size),[186] and (2) product type (discrete, memory or logic).[187] For each linewidth technology, a weighted average number of layers is estimated using VLSI product-specific worldwide silicon demand data in conjunction with complexity factors (i.e., the number of layers per Integrated Circuit (IC)) specific to product type (Burton and Beizaie 2001, ITRS 2007). PEVM derives historical consumption of silicon (i.e., square inches) by linewidth technology from published data on annual wafer starts and average wafer size (VLSI Research, Inc. 2012).

The emission factor in PEVM is the average of four historical emission factors, each derived by dividing the total annual emissions reported by the Partners for each of the four years between 1996 and 1999 by the total TMLA estimated for the Partners in each of those years. Over this period, the emission factors varied relatively little (i.e., the relative standard deviation for the average was 5 percent). Since Partners are believed not to have applied significant emission reduction measures before 2000, the resulting average emission factor reflects uncontrolled emissions. The emission factor is used to estimate world uncontrolled emissions using publicly-available data on world silicon consumption.

As it was assumed for this time period that there was no consequential adoption of fluorinated-gas-reducing measures, a fixed distribution of fluorinated-gas use was assumed to apply to the entire U.S. industry to estimate gas-specific emissions. This distribution was based upon the average fluorinated-gas purchases made by semiconductor manufacturers during this period and the application of IPCC default emission factors for each gas (Burton and Beizaie 2001).

To estimate N_2O emissions, it is assumed the proportion of N_2O emissions estimated for 1995 (discussed below) remained constant for the period of 1990 through1994.

1995 through 1999

For 1995 through 1999, total U.S. emissions were extrapolated from the total annual emissions reported by the Partners (1995 through 1999). Partner-reported emissions are considered more representative (e.g., in terms of capacity utilization in a given year) than PEVM estimated emissions, and are used to generate total U.S. emissions when applicable. The emissions reported by the Partners were divided by the ratio of the total capacity of the plants operated by the Partners and the total capacity of all of the semiconductor plants in the United States; this ratio represents the share of capacity attributable to the Partnership. This method assumes that Partners and non-Partners have identical capacity utilizations and distributions of manufacturing technologies. Plant capacity data is contained in the World Fab Forecast (WFF) database and its predecessors, which is updated quarterly (Semiconductor Equipment and Materials Industry 2012 and 2013). Gas-specific emissions were estimated using the same method as for 1990 through 1994.

For this time period, the N_2O emissions were estimated using an emission factor that is applied to the annual, total U.S. TMLA manufactured. The emission factor was developed using a regression-through-the-origin (RTO) model: GHGRP reported N_2O emissions were regressed against the corresponding TMLA of facilities that reported no use of abatement systems. Details on the GHGRP reported emissions and development of emission factor using the RTO model are presented in the 2011 through 2013 section. The total U.S. TMLA manufactured were estimated using PEVM.

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[186] By decreasing features of Integrated Circuit components, more components can be manufactured per device, which increases its functionality. However, as those individual components shrink it requires more layers to interconnect them to achieve the functionality. For example, a microprocessor manufactured with 65 nm feature sizes might contain as many as 1 billion transistors and require as many as 11 layers of component interconnects to achieve functionality, while a device manufactured with 130 nm feature size might contain a few hundred million transistors and require 8 layers of component interconnects (ITRS 2007).

[187] Memory devices manufactured with the same feature sizes as microprocessors (a logic device) require approximately one-half the number of interconnect layers, whereas discrete devices require only a silicon base layer and no interconnect layers (ITRS 2007). Since discrete devices did not start using PFCs appreciably until 2004, they are only accounted for in the PEVM emissions estimates from 2004 onwards.

2000 through 2006

Emissions for the years 2000 through 2006—the period during which Partners began the consequential application of fluorinated greenhouse gas-reduction measures—were estimated using a combination of Partner-reported emissions and adjusted PEVM modeled emissions. The emissions reported by Partners for each year were accepted as the quantity emitted from the share of the industry represented by those Partners. Remaining emissions, those from non-Partners, were estimated using PEVM, with one change. To ensure time series consistency and to reflect the increasing use of remote clean technology (which increases the efficiency of the production process while lowering emissions of fluorinated greenhouse gases), the average non-Partner emission factor was assumed to begin declining gradually during this period. Specifically, the non-Partner emission factor for each year was determined by linear interpolation, using the end points of 1999 (the original PEVM emission factor) and 2011 (a new emission factor determined for the non-Partner population based on GHGRP-reported data, described below).

The portion of the U.S. total attributed to non-Partners is obtained by multiplying PEVM's total U.S. emissions figure by the non-Partner share of U.S. total silicon capacity for each year as described above.[188] Gas-specific emissions from non-Partners were estimated using linear interpolation of gas-specific emission distribution of 1999 (assumed same as total U.S. Industry in 1994) and 2011 (calculated from a subset of non-Partner facilities from GHGRP reported emissions data). Annual updates to PEVM reflect published figures for actual silicon consumption from VLSI Research, Inc., revisions and additions to the world population of semiconductor manufacturing plants, and changes in IC fabrication practices within the semiconductor industry (see ITRS 2008 and Semiconductor Equipment and Materials Industry 2011).[189,190,191]

The N_2O emissions were estimated using the same methodology as 1995-1999 methodology.

2007 through 2010

For the years 2007 through 2010, emissions were also estimated using a combination of Partner reported emissions and adjusted PEVM modeled emissions to provide estimates for non-Partners; however, two improvements were made to the estimation method employed for the previous years in the time series. First, the 2007 through 2010 emission estimates account for the fact that Partners and non-Partners employ different distributions of manufacturing technologies, with the Partners using manufacturing technologies with greater transistor densities and

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[188] This approach assumes that the distribution of linewidth technologies is the same between Partners and non-Partners. As discussed in the description of the method used to estimate 2007 emissions, this is not always the case.

[189] Special attention was given to the manufacturing capacity of plants that use wafers with 300 mm diameters because the actual capacity of these plants is ramped up to design capacity, typically over a 2–3 year period. To prevent overstating estimates of partner-capacity shares from plants using 300 mm wafers, *design* capacities contained in WFW were replaced with estimates of *actual installed* capacities for 2004 published by Citigroup Smith Barney (2005). Without this correction, the partner share of capacity would be overstated, by approximately 5 percent. For perspective, approximately 95 percent of all new capacity additions in 2004 used 300 mm wafers, and by year-end those plants, on average, could operate at approximately 70 percent of the design capacity. For 2005, actual installed capacities were estimated using an entry in the World Fab Watch database (April 2006 Edition) called "wafers/month, 8-inch equivalent," which denoted the actual installed capacity instead of the fully-ramped capacity. For 2006, actual installed capacities of new fabs were estimated using an average monthly ramp rate of 1100 wafer starts per month (wspm) derived from various sources such as semiconductor fabtech, industry analysts, and articles in the trade press. The monthly ramp rate was applied from the first-quarter of silicon volume (FQSV) to determine the average design capacity over the 2006 period.

[190] In 2006, the industry trend in co-ownership of manufacturing facilities continued. Several manufacturers, who are Partners, now operate fabs with other manufacturers, who in some cases are also Partners and in other cases are not Partners. Special attention was given to this occurrence when estimating the Partner and non-Partner shares of U.S. manufacturing capacity.

[191] Two versions of PEVM are used to model non-Partner emissions during this period. For the years 2000 to 2003 PEVM v3.2.0506.0507 was used to estimate non-Partner emissions. During this time, discrete devices did not use PFCs during manufacturing and therefore only memory and logic devices were modeled in the PEVM v3.2.0506.0507. From 2004 onwards, discrete device fabrication started to use PFCs, hence PEVM v4.0.0701.0701, the first version of PEVM to account for PFC emissions from discrete devices, was used to estimate non-Partner emissions for this time period.

therefore greater numbers of layers.[192] Second, the scope of the 2007 through 2010 estimates was expanded relative to the estimates for the years 2000 through 2006 to include emissions from research and development (R&D) fabs. This additional enhancement was feasible through the use of more detailed data published in the WFF. PEVM databases were updated annually as described above. The published world average capacity utilization for 2007 through 2010 was used for production fabs, while for R&D fabs a 20 percent figure was assumed (SIA 2009).

In addition, publicly-available actual utilization data was used to account for differences in fab utilization for manufacturers of discrete and IC products for 2010 emissions for non-Partners. PEVM estimates were adjusted using technology-weighted capacity shares that reflect the relative influence of different utilization. Gas-specific emissions for non-Partners were estimated using the same method as for 2000 through 2006.

The N_2O emissions were estimated using the same methodology as 1995 through 1999 methodology.

2011 through 2013

The fifth and final method for estimating emissions from semiconductor manufacturing covers the period 2011 through 2013, the years after EPA's Partnership with the semiconductor industry ended (in 2010) and reporting under the GHGRP began. Manufacturers whose estimated uncontrolled emissions equal or exceed 25,000 mt CO_2 Eq. per year (based on default emission factors and total capacity in terms of substrate area) are required to report their emissions to the EPA. This population of reporters to EPA's GHGRP included both historical Partners of EPA's PFC Reduction/Climate Partnership as well as non-Partners. In EPA's GHGRP, the population of non-Partner facilities also included manufacturers that use GaAs technology in addition to Si technology[193]. Emissions from the population of manufacturers that were below the reporting threshold were also estimated for this time period using EPA-developed emission factors and estimates of facility-specific production obtained from WFF. Inventory totals reflect the emissions from both populations.

Under EPA's GHGRP, semiconductor manufacturing facilities report emissions of fluorinated GHGs used in etch and clean processes and as heat transfer fluids. They also report N_2O emissions from CVD and other processes. The fluorinated GHGs, and N_2O were aggregated, by gas, across all semiconductor manufacturing GHGRP reporters to calculate gas-specific emissions for the GHGRP-reporting segment of the U.S. industry.

For the segment of the semiconductor industry, which is below EPA's GHGRP reporting threshold, and for R&D facilities, which are not covered by EPA's GHGRP, emission estimates are based on EPA-developed emission factors for the fluorinated GHGs and N_2O. The new emission factors (in units of mass of CO_2 Eq. / TMLA [MSI]) are based on the emissions reported by facilities under EPA's GHGRP and TMLA estimates for these facilities from the WFF (SEMI 2012 and SEMI 2013). In a refinement of the method used in prior years to estimate emissions for the non-Partner population, different emission factors were developed for different subpopulations of fabs, one for facilities that manufacture devices on Si wafers and one for facilities that manufacture on GaAs wafers. An analysis of the emission factors of reporting fabs showed that the characteristics that had the largest impacts on emission factors were the substrate (i.e., Si or GaAs) used at the fab, whether the fab contained R&D activities, and whether the fab reported using point-of-use fluorinated greenhouse gas abatement[194]. For each of these groups, a subpopulation-specific emission factor was obtained using a regression-through-the-origin (RTO) model: facility-reported aggregate emissions of seven fluorinated GHGs (CF_4, C_2F_6, C_3F_8, C_4F_8, CHF_3, SF_6 and NF_3)[195] were regressed against the corresponding TMLA to estimate an aggregate F-GHG emissions factor (CO_2 Eq./MSI TMLA)

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[192] EPA considered applying this change to years before 2007, but found that it would be difficult due to the large amount of data (i.e., technology-specific global and non-Partner TMLA) that would have to be examined and manipulated for each year. This effort did not appear to be justified given the relatively small impact of the improvement on the total estimate for 2007 and the fact that the impact of the improvement would likely be lower for earlier years because the estimated share of emissions accounted for by non-Partners is growing as Partners continue to implement emission-reduction efforts.

[193] GaAs and Si technologies refer to the wafer on which devices are manufactured, which use the same PFCs but in different ways.

[194] For the non-reporting segment of the industry using GaAs technology, emissions were estimated only for those fabs that manufactured the same products as manufactured by reporters. The products manufactured were categorized as discrete (emissions did not scale up with decreasing feature size).

[195] Only seven gases were aggregated because inclusion of fluorinated GHGs that are not reported in the inventory results in overestimation of emission factor that is applied to the various non-reporting subpopulations.

and facility-reported N_2O emissions were regressed against the corresponding TMLA to estimate a N_2O emissions factor (CO_2 Eq./MSI TMLA). For each subpopulation, the slope of the RTO model is the emission factor for that subpopulation. To estimate emissions from fabs that are solely doing research and development (R&D) or are Pilot fabs (i.e., fabs that are excluded from subpart I reporting requirements), emission factors were estimated based on GHGRP reporting fabs containing R&D activities. EPA applied a scaling factor of 1.15 to the slope of the RTO model to estimate the emission factor applicable to the non-reporting fabs that are only R&D or Pilot fabs. This was done as R&D activities lead to use of more F-GHGs and N_2O for development of chips that are not counted towards the final estimated TMLA. Hence, it is assumed that the fabs with only R&D activities use 15 percent more F-GHGs and N_2O per TMLA. However, as was assumed for 2007 through 2010, fabs with only R&D activities were assumed to utilize only 20 percent of their manufacturing capacity. Other fabs were assumed to utilize 89 percent of their manufacturing capacity, held constant at 2012 levels which is slightly lower than 2011 levels. Fabs that produce discrete products are assumed to utilize 84 percent of their manufacturing capacity, held constant at 2011 levels. These utilizations at 2011 levels are based on the Semiconductor Industry Association report (SICAS, 2011).

Non-reporting fabs were then broken out into similar subpopulations. Information on the technology and R&D activities of non-reporting fabs was available through the WFF. Information on the use of point-of-use abatement by non-reporting fabs was not available; thus, EPA conservatively assumed that non-reporting facilities did not use point-of-use abatement. The appropriate emission factor was applied to the total TMLA of each subpopulation of non-reporting facilities to estimate the GWP-weighted emissions of that subpopulation.

Gas-specific, GWP-weighted emissions for each subpopulation of non-reporting facilities were estimated using the corresponding reported distribution of gas-specific, GWP-weighted emissions from which the aggregate emission factors were developed. Estimated in this manner, the non-reporting population accounted for 9, 10 and 10 percent of U.S. emissions in 2011, 2012 and 2013, respectively. The GHGRP-reported emissions and the calculated non-reporting population emissions are summed to estimate the total emissions from semiconductor manufacturing.

The methodology used for this time period included, for the first time, emissions from facilities employing Si- and GaAs-using technologies. The use of GaAs technology became evident via analysis of GHGRP emissions and WFF data. However, no adjustment of pre-2011 emissions was made because (1) the use of these technologies appears relatively new, (2) in the aggregate make a relatively small contribution to total industry emissions (i.e., 4 percent in 2013), and (3) would require a large effort to retroactively adjust pre-2011 emissions.

Data Sources

GHGRP reporters estimated their emissions using a default emission factor method established by EPA. This method is very similar to the Tier 2b Method in the *2006 IPCC Guidelines*, but it goes beyond that method by establishing different default emission and by-product generation factors for different wafer sizes (i.e., 300mm vs. 150 and 200mm) and CVD clean subtypes (in situ thermal, in situ thermal, and remote plasma). Partners estimated their emissions using a range of methods. It is assumed that most Partners used a method at least as accurate as the IPCC's Tier 2a Methodology, recommended in the *2006 IPCC Guidelines*. Estimates of operating plant capacities and characteristics for Partners and non-Partners were derived from the Semiconductor Equipment and Materials Industry (SEMI) WFF (formerly World Fab Watch) database (1996 through 2013) (e.g., Semiconductor Materials and Equipment Industry, 2013). Actual worldwide capacity utilizations for 2011 were obtained from Semiconductor International Capacity Statistics (SICAS) (SIA, 2011). Estimates of the number of layers for each linewidth was obtained from International Technology Roadmap for Semiconductors: 2013 Edition (Burton and Beizaie 2001, ITRS 2007, ITRS 2008, ITRS 2011, ITRS 2013). PEVM utilized the WFF, SICAS, and ITRS, as well as historical silicon consumption estimates published by VLSI.

Uncertainty and Time-Series Consistency

A quantitative uncertainty analysis of this source category was performed using the IPCC-recommended Approach 2 uncertainty estimation methodology, the Monte Carlo Stochastic Simulation technique. The equation used to estimate uncertainty is:

$$\text{Total Emissions } (E_T) = \text{GHGRP Reported F-GHG Emissions } (E_{R,F\text{-}GHG}) + \text{Non-Reporters' Estimated F-GHG Emissions } (E_{NR,F\text{-}GHG}) + \text{GHGRP Reported } N_2O \text{ Emissions } (E_{R,N_2O}) + \text{Non-Reporters' Estimated } N_2O \text{ Emissions } (E_{NR,N_2O})$$

where E_R and E_{NR} denote totals for the indicated subcategories of emissions for F-GHG and N_2O, respectively.

The uncertainty in E_T presented in Table 4-93 below results from the convolution of four distributions of emissions, each reflecting separate estimates of possible values of $E_{R,F\text{-}GHG}$, E_{R,N_2O}, $E_{NR,F\text{-}GHG}$, and E_{NR,N_2O}. The approach and methods for estimating each distribution and combining them to arrive at the reported 95 percent CI are described in the remainder of this section.

The uncertainty estimate of $E_{R,\ F\text{-}GHG}$, or GHGRP reported F-GHG emissions, is developed based on gas-specific uncertainty estimates of emissions for two industry segments, one processing 200 mm wafers and one processing 300 mm wafers. Uncertainties in emissions for each gas and industry segment were developed during the assessment of emission estimation methods for the subpart I GHGRP rulemaking in 2012 (see *Technical Support for Modifications to the Fluorinated Greenhouse Gas Emission Estimation Method Option for Semiconductor Facilities under Subpart* I, docket EPA–HQ–OAR–2011–0028).[196] The 2012 analysis did not take into account the use of abatement. For the industry segment that processed 200 mm wafers, estimates of uncertainties at a 95 percent CI ranged from ±29 percent for C_3F_8 to ±10 percent for CF_4. For the corresponding 300 mm industry segment, estimates of the 95 percent CI ranged from ±36 percent for C_4F_8 to ±16 percent for CF_4. These gas and wafer-specific uncertainty estimates are applied to the total emissions of the facilities that did not abate emissions as reported under EPA's GHGRP.

For those facilities reporting abatement of emissions under EPA's GHGRP, estimates of uncertainties for the no abatement industry segments are modified to reflect the use of full abatement (abatement of *all* gases from *all* cleaning and etching equipment) and partial abatement. These assumptions used to develop uncertainties for the partial and full abatement facilities are identical for 200 mm and 300 mm wafer processing facilities. For all facilities reporting gas abatement, a triangular distribution of destruction or removal efficiency is assumed for each gas. The triangular distributions range from an asymmetric and highly uncertain distribution of 0 percent minimum to 90 percent maximum with 70 percent most likely value for CF_4 to a symmetric and less uncertain distribution of 85 percent minimum to 95 percent maximum with 90 percent most likely value for C_4F_8, NF_3 and SF_6. For facilities reporting partial abatement, the distribution of fraction of the gas fed through the abatement device, for each gas, is assumed to be triangularly distributed as well. It is assumed that no more than 50 percent of the gases area abated (i.e., the maximum value) and that 50 percent is the most likely value and the minimum is 0 percent. Consideration of abatement then resulted in four additional industry segments, two 200 mm wafer-processing segments (one fully and one partially abating each gas) and two 300 mm wafer-processing segment (one fully and the other partially abating each gas). Gas-specific emission uncertainties were estimated by convolving the distributions of unabated emissions with the appropriate distribution of abatement efficiency for fully and partially abated facilities using a Montel Carlo simulation.

The uncertainty in $E_{R,F\text{-}GHG}$ is obtained by allocating the estimates of uncertainties to the total GHGRP-reported emissions from each of the six industry segments, and then running a Monte Carlo simulation which results in the 95 percent CI for emissions from GHGRP reporting facilities ($E_{R,F\text{-}GHG}$).

The uncertainty in $E_{R,N2O}$ is obtained by assuming that the uncertainty in the emissions reported by each of the GHGRP reporting facilities results from the uncertainty in quantity of N_2O consumed and the N_2O emission factor (or utilization). Similar to analyses completed for subpart I (see *Technical Support for Modifications to the Fluorinated Greenhouse Gas Emission Estimation Method Option for Semiconductor Facilities under Subpart* I, docket EPA–HQ–OAR–2011–0028), the uncertainty of N_2O consumed was assumed to be 20 percent. Consumption of N_2O for GHGRP reporting facilities was estimated by back- calculating from emissions reported and assuming no abatement. The quantity of N_2O utilized (the complement of the emission factor) was assumed to have a triangular

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[196] On November 13, 2013, EPA published a final rule revising subpart I (Electronics Manufacturing) of the GHGRP (78 FR 68162). The revised rule includes updated default emission factors and updated default destruction and removal efficiencies that are slightly different from those that semiconductor manufacturers were required to use to report their 2012 emissions. The uncertainty analyses that were performed during the development of the revised rule focused on these updated defaults, but are expected to be reasonably representative of the uncertainties associated with the older defaults, particularly for estimates at the country level. (They may somewhat underestimate the uncertainties associated with the older defaults at the facility level.) For simplicity, the 2012 estimates are assumed to be unbiased although in some cases, the updated (and therefore more representative) defaults are higher or lower than the older defaults. Multiple models and sensitivity scenarios were run for the subpart I analysis. The uncertainty analysis presented here made use of the Input gas and wafer size model (Model 1) under the following conditions: Year = 2010, f = 20, n = SIA3.

distribution with a minimum value of 0 percent, mode of 20 percent and maximum value of 84 percent. The minimum was selected based on physical limitations, the mode was set equivalent to the subpart I default N_2O utilization rate for chemical vapor deposition, and the maximum was set equal to the maximum utilization rate found in ISMI Analysis of Nitrous Oxide Survey Data (ISMI, 2009). The inputs were used to simulate emissions for each of the GHGRP reporting, N_2O-emitting facilities. The uncertainty for the total reported N_2O emissions was then estimated by combining the uncertainties of each of the facilities reported emissions using Monte Carlo simulation.

The estimate of uncertainty in $E_{NR,F\text{-}GHG}$ and $E_{NR,N2O}$ entailed developing estimates of uncertainties for the emissions factors for each non-reporting sub-category and the corresponding estimates of TMLA.

The uncertainty in TMLA depends on the uncertainty of two variables—an estimate of the uncertainty in the average annual capacity utilization for each level of production of fabs (e.g., full scale or R&D production) and a corresponding estimate of the uncertainty in the number of layers manufactured. For both variables, the distributions of capacity utilizations and number of manufactured layers are assumed triangular for all categories of non-reporting fabs. For production fabs the most probable utilization is assumed to be 89 percent, with the highest and lowest utilization assumed to be 100 percent and 63 percent, respectively. The corresponding values for facilities that manufacture discrete devices are, 84 percent, 100 percent, and 66 percent, respectively, while the values for utilization for R&D facilities, are assumed to be 20 percent, 33 percent, and 9 percent, respectively. The most probable utilizations are unchanged compared to 2012 Inventory year. To address the uncertainty in the capacity utilization for Inventory year 2013, the lower bound has been decreased by 10 percent, and the upper bound has been increased by 10 percent (or 100 percent if greater than 100 percent) compared to the bounds used in the 2012 Inventory year. For the triangular distributions that govern the number of possible layers manufactured, it is assumed the most probable value is one layer less than reported in the ITRS; the smallest number varied by technology generation between one and two layers less than given in the ITRS and largest number of layers corresponded to the figure given in the ITRS.

The uncertainty bounds for the average capacity utilization and the number of layers manufactured are used as inputs in a separate Monte Carlo simulation to estimate the uncertainty around the TMLA of both individual facilities as well as the total non-reporting TMLA of each sub-population.

The uncertainty around the emission factors for each non-reporting category of facilities is dependent on the uncertainty of the total emissions (MMT CO_2 Eq. units) and the TMLA of each reporting facility in that category. For each subpopulation of reporting facilities, total emissions were regressed on TMLA (with an intercept forced to zero) for 10,000 emissions and 10,000 TMLA values in a Monte Carlo simulation, which results in 10,000 total regression coefficients (emission factors). The 2.5th and the 97.5th percentile of these emission factors are determined and the bounds are assigned as the percent difference from the estimated emission factor.

For simplicity, the results of the Monte Carlo simulations on the bounds of the gas- and wafer size-specific emissions as well as the TMLA and emission factors are assumed to be normally distributed and the uncertainty bounds are assigned at 1.96 standard deviations around the estimated mean. The departures from normality were observed to be small.

The final step in estimating the uncertainty in emissions of non-reporting facilities is convolving the distribution of emission factors with the distribution of TMLA using Monte Carlo simulation.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-93, which is also obtained by convolving—using Monte Carlo simulation—the distributions of emissions for each reporting and non-reporting facility. The emissions estimate for total U.S. F-GHG and N_2O emissions from semiconductor manufacturing were estimated to be between 4.0 and 4.4 MMT CO_2 Eq. at a 95 percent confidence level. This range represents 5 percent below to 5 percent above the 2013 emission estimate of 4.2 MMT CO_2 Eq. This range and the associated percentages apply to the estimate of total emissions rather than those of individual gases. Uncertainties associated with individual gases will be somewhat higher than the aggregate, but were not explicitly modeled.

Table 4-93: Approach 2 Quantitative Uncertainty Estimates for HFC, PFC, SF_6, NF_3 and N_2O Emissions from Semiconductor Manufacture (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound[b]	Upper Bound[b]	Lower Bound	Upper Bound
Semiconductor Manufacture	HFC, PFC, SF_6, NF_3, and N_2O	4.2	4.0	4.4	-5%	5%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

[b] Absolute lower and upper bounds were calculated using the corresponding lower and upper bounds in percentages.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in emissions from CH_4, HFCs, PFCs, SF_6, and NF_3. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

The decrease in the GWP of SF_6 and increase in the GWP of all other gases had several impacts on Inventory estimates. In the 1990 through 1994 time period, an overall increase in total annual GWP-weighted emissions is seen. In the 1995 through 2010 time period, the Inventory methodology relies on various gas distributions based on Partner reported emissions and PEVM estimated emissions. The changes in GWP carry through to changes in the estimated gas distributions, and hence changes in gas-by-gas emission estimates, in CO_2 Eq., and total annual fluorinated greenhouse gas emission estimates, in CO_2 Eq..

For the first time, NF_3 and N_2O have been included in total annual GWP-weighted emission estimates for the United States. This, along with an increased weighted GWP from SAR to AR4 led to increase in total emissions for all years as compared to previous Inventories. The emissions of each gas were impacted by the increase in overall emissions as well as the percent distribution of each gas as a result of changes in their GWPs.

Emissions in years 2011 and 2012 were updated to reflect updated emissions reporting in EPA's GHGRP. For the non-reporting population, the methodology to determine the non-reporting population for GaAs using facilities has been updated. In the updated methodology, revised assumptions were made about the GaAs using facilities that use fluorinated greenhouse gases (e.g., only the non-reporters that use wafers greater than or equal to four inches have been assumed to use fluorinated greenhouse gases, facilities that use wafers less than 4 inches are assumed to use wet etching and hence do not consume or emit any fluorinated greenhouse gases). Further, EPA has drawn an analogy between GaAs-using GHGRP reporters and non-reporters provided the non-reporters use wafers greater than 4 inches and manufacture the many versions of high electron mobility transistors (HEMT, PHEMT, MHEMT, HET, MOFETs), which are discrete devices and may be made to specific order by certain foundries. By virtue of this analogy, EPA has estimated emissions only from the non-reporters that use GaAs technology and manufacture HEMT and their variations. While other devices may be made using GaAs technology, EPA has no reporters under the GHGRP that manufacture them and hence has no basis for estimating an emission factor. EPA has thus assumed that they do not use or emit F-GHGs. This has decreased the non-reporting facilities subpopulation, and subsequently total emissions for the years 2011 and 2012.

Planned Improvements

This Inventory contains estimates of seven fluorinated gases for semiconductor manufacturing. However, other fluorinated gases (e.g., C_5F_8) are used in relatively smaller, but significant amounts. Previously, emissions data for these other fluorinated gases was not reported through the EPA Partnership. Through EPA's GHGRP, these data, as well as heat transfer fluid emission data, are available. Therefore, a point of consideration for future Inventory reports is the inclusion of other fluorinated gases, and emissions from heat transfer fluid (HTF) loss to the atmosphere.

Fluorinated heat transfer fluids, of which some are liquid perfluorinated compounds, are used for temperature control, device testing, cleaning substrate surfaces and other parts, and soldering in certain types of semiconductor manufacturing production processes. Evaporation of these fluids is a source of fluorinated emissions (EPA 2006). The GHGRP-reported HTF emissions along with WFF database could be used to develop emission factors for identified subpopulations. Further research needs to be done to determine if the same subpopulations identified in developing new emission factors for F-GHGs are applicable or new subpopulations have to be studied as HTFs are used primarily by manufacturers of wafer size 300 mm and above.

Along with more emissions information for semiconductor manufacturing, EPA's GHGRP requires the reporting of emissions from other types of electronics manufacturing, including micro-electro-mechanical systems, flat panel displays, and photovoltaic cells. There currently are no flat panel displays, and photovoltaic cell manufacturing facilities that are reporting to EPA's GHGRP, and five reporting MEMs manufacturers. The MEMs manufacturers also report emissions from semiconductor manufacturing and do not distinguish between these two types of manufacturing in their report; thus, emissions from MEMs manufacturers are included in the totals here. Emissions from manufacturing of flat panel displays and photovoltaic cells may be included in future Inventory reports; however, estimation methodologies would need to be developed.

4.23 Substitution of Ozone Depleting Substances (IPCC Source Category 2F)

Hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) are used as alternatives to several classes of ozone-depleting substances (ODSs) that are being phased out under the terms of the *Montreal Protocol* and the Clean Air Act Amendments of 1990.[197] Ozone depleting substances—chlorofluorocarbons (CFCs), halons, carbon tetrachloride, methyl chloroform, and hydrochlorofluorocarbons (HCFCs)—are used in a variety of industrial applications including refrigeration and air conditioning equipment, solvent cleaning, foam production, sterilization, fire extinguishing, and aerosols. Although HFCs and PFCs are not harmful to the stratospheric ozone layer, they are potent greenhouse gases. Emission estimates for HFCs and PFCs used as substitutes for ODSs are provided in Table 4-94 and Table 4-95.

Table 4-94: Emissions of HFCs and PFCs from ODS Substitutes (MMT CO_2 Eq.)

Gas	1990	2005	2009	2010	2011	2012	2013
HFC-23	0.0	+	+	+	+	+	+
HFC-32	0.0	0.3	1.8	2.6	3.3	4.3	5.2
HFC-125	+	11.0	22.0	28.1	33.7	40.0	46.3
HFC-134a	+	81.9	87.9	86.5	81.4	76.5	71.3
HFC-143a	+	10.7	15.5	17.9	20.3	22.8	25.3
HFC-236fa	0.0	1.2	1.4	1.4	1.4	1.5	1.5
CF_4	0.0	+	+	+	+	+	+
Others*	0.3	5.9	7.4	7.8	8.2	8.6	9.0
Total	**0.3**	**111.1**	**136.0**	**144.4**	**148.4**	**153.5**	**158.6**

+ Does not exceed 0.05 MMT CO_2 Eq.

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[197] [42 U.S.C § 7671, CAA Title VI]

* Others include HFC-152a, HFC-227ea, HFC-245fa, HFC-43-10mee, C_4F_{10}, and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications. For estimating purposes, the GWP value used for PFC/PFPEs was based upon C_6F_{14}.
Note: Totals may not sum due to independent rounding.

Table 4-95: Emissions of HFCs and PFCs from ODS Substitution (MT)

Gas	1990	2005	2009	2010	2011	2012	2013
HFC-23	+	1	2	2	2	2	2
HFC-32	+	505	2,611	3,849	4,925	6,309	7,733
HFC-125	+	3,147	6,290	8,038	9,615	11,415	13,236
HFC-134a	+	57,286	61,467	60,509	56,929	53,478	49,837
HFC-143a	+	2,401	3,460	3,996	4,547	5,091	5,651
HFC-236fa	+	125	144	146	147	148	151
CF_4	+	2	3	3	4	4	4
Others*	M	M	M	M	M	M	M

M (Mixture of Gases)
+ Does not exceed 0.5 MT
* Others include HFC-152a, HFC-227ea, HFC-245fa, HFC-43-10mee, C_4F_{10}, and PFC/PFPEs, the latter being a proxy for a diverse collection of PFCs and perfluoropolyethers (PFPEs) employed for solvent applications.

In 1990 and 1991, the only significant emissions of HFCs and PFCs as substitutes to ODSs were relatively small amounts of HFC-152a—used as an aerosol propellant and also a component of the refrigerant blend R-500 used in chillers—and HFC-134a in refrigeration end-uses. Beginning in 1992, HFC-134a was used in growing amounts as a refrigerant in motor vehicle air-conditioners and in refrigerant blends such as R-404A.[198] In 1993, the use of HFCs in foam production began, and in 1994 ODS substitutes for halons entered widespread use in the United States as halon production was phased-out. In 1995, these compounds also found applications as solvents.

The use and subsequent emissions of HFCs and PFCs as ODS substitutes has been increasing from small amounts in 1990 to 158.6 MMT CO_2 Eq. in 2013. This increase was in large part the result of efforts to phase out CFCs and other ODSs in the United States. In the short term, this trend is expected to continue, and will likely continue over the next decade as HCFCs, which are interim substitutes in many applications, are themselves phased-out under the provisions of the Copenhagen Amendments to the *Montreal Protocol*. Improvements in the technologies associated with the use of these gases and the introduction of alternative gases and technologies, however, may help to offset this anticipated increase in emissions.

Table 4-96 presents emissions of HFCs and PFCs as ODS substitutes by end-use sector for 1990 through 2013. The end-use sectors that contributed the most toward emissions of HFCs and PFCs as ODS substitutes in 2013 include refrigeration and air-conditioning (137.6 MMT CO_2 Eq., or approximately 87 percent), aerosols (10.5 MMT CO_2 Eq., or approximately 7 percent), and foams (7.4 MMT CO_2 Eq., or approximately 5 percent). Within the refrigeration and air-conditioning end-use sector, motor vehicle air-conditioning was the highest emitting end-use (44.1 MMT CO_2 Eq.), followed by refrigerated retail food and refrigerated transport. Each of the end-use sectors is described in more detail below.

Table 4-96: Emissions of HFCs and PFCs from ODS Substitutes (MMT CO_2 Eq.) by Sector

Sector	1990	2005	2009	2010	2011	2012	2013
Refrigeration/Air Conditioning	+	99.2	119.7	126.0	129.0	133.3	137.6
Aerosols	0.3	7.6	9.4	9.7	10.1	10.3	10.5
Foams	+	2.1	4.2	5.9	6.4	6.9	7.4
Solvents	+	1.7	1.6	1.7	1.7	1.7	1.8
Fire Protection	+	0.7	1.0	1.1	1.2	1.3	1.3
Total	**0.3**	**111.1**	**136.0**	**144.4**	**148.4**	**153.5**	**158.6**

+ Does not exceed 0.05 MMT CO_2 Eq.

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[198] R-404A contains HFC-125, HFC-143a, and HFC-134a.

Refrigeration/Air Conditioning

The refrigeration and air-conditioning sector includes a wide variety of equipment types that have historically used CFCs or HCFCs. End-uses within this sector include motor vehicle air-conditioning, retail food refrigeration, refrigerated transport (e.g., ship holds, truck trailers, railway freight cars), household refrigeration, residential and small commercial air-conditioning and heat pumps, chillers (large comfort cooling), cold storage facilities, and industrial process refrigeration (e.g., systems used in food processing, chemical, petrochemical, pharmaceutical, oil and gas, and metallurgical industries). As the ODS phaseout is taking effect, most equipment is being or will eventually be retrofitted or replaced to use HFC-based substitutes. Common HFCs in use today in refrigeration/air-conditioning equipment are HFC-134a, R-410A,[199] R-404A, and R-507A.[200] These HFCs are emitted to the atmosphere during equipment manufacture and operation (as a result of component failure, leaks, and purges), as well as at servicing and disposal events.

Aerosols

Aerosol propellants are used in metered dose inhalers (MDIs) and a variety of personal care products and technical/specialty products (e.g., duster sprays and safety horns). Many pharmaceutical companies that produce MDIs—a type of inhaled therapy used to treat asthma and chronic obstructive pulmonary disease—have replaced the use of CFCs with HFC-propellant alternatives. The earliest ozone-friendly MDIs were produced with HFC-134a, but the industry has started to use HFC-227ea as well. Conversely, since the use of CFC propellants was banned in 1978, most non-medical consumer aerosol products have not transitioned to HFCs, but to “not-in-kind” technologies, such as solid roll-on deodorants and finger-pump sprays. The transition away from ODS in specialty aerosol products has also led to the introduction of non-fluorocarbon alternatives (e.g., hydrocarbon propellants) in certain applications, in addition to HFC-134a or HFC-152a. These propellants are released into the atmosphere as the aerosol products are used.

Foams

CFCs and HCFCs have traditionally been used as foam blowing agents to produce polyurethane (PU), polystyrene, polyolefin, and phenolic foams, which are used in a wide variety of products and applications. Since the *Montreal Protocol*, flexible PU foams as well as other types of foam, such as polystyrene sheet, polyolefin, and phenolic foam, have transitioned almost completely away from fluorocompounds, into alternatives such as CO_2, methylene chloride, and hydrocarbons. The majority of rigid PU foams have transitioned to HFCs—primarily HFC-134a and HFC-245fa. Today, these HFCs are used to produce polyurethane appliance, PU commercial refrigeration, PU spray, and PU panel foams—used in refrigerators, vending machines, roofing, wall insulation, garage doors, and cold storage applications. In addition, HFC-152a, HFC-134a and CO_2 are used to produce polystyrene sheet/board foam, which is used in food packaging and building insulation. Emissions of blowing agents occur when the foam is manufactured as well as during the foam lifetime and at foam disposal, depending on the particular foam type.

Solvents

CFCs, methyl chloroform (1,1,1-trichloroethane or TCA), and to a lesser extent carbon tetrachloride (CCl_4) were historically used as solvents in a wide range of cleaning applications, including precision, electronics, and metal cleaning. Since their phaseout, metal cleaning end-use applications have primarily transitioned to non-fluorocarbon solvents and not-in-kind processes. The precision and electronics cleaning end-uses have transitioned in part to high-GWP gases, due to their high reliability, excellent compatibility, good stability, low toxicity, and selective solvency. These applications rely on HFC-43-10mee, HFC-365mfc, HFC-245fa, and to a lesser extent, PFCs. Electronics cleaning involves removing flux residue that remains after a soldering operation for printed circuit boards and other contamination-sensitive electronics applications. Precision cleaning may apply to either electronic components or to metal surfaces, and is characterized by products, such as disk drives, gyroscopes, and optical components, that

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[199] R-410A contains HFC-32 and HFC-125.

[200] R-507A, also called R-507, contains HFC-125 and HFC-143a.

require a high level of cleanliness and generally have complex shapes, small clearances, and other cleaning challenges. The use of solvents yields fugitive emissions of these HFCs and PFCs.

Fire Protection

Fire protection applications include portable fire extinguishers (“streaming” applications) that originally used halon 1211, and total flooding applications that originally used halon 1301, as well as some halon 2402. Since the production and sale of halons were banned in the United States in 1994, the halon replacement agent of choice in the streaming sector has been dry chemical, although HFC-236fa is also used to a limited extent. In the total flooding sector, HFC-227ea has emerged as the primary replacement for halon 1301 in applications that require clean agents. Other HFCs, such as HFC-23 and HFC-125, are used in smaller amounts. The majority of HFC-227ea in total flooding systems is used to protect essential electronics, as well as in civil aviation, military mobile weapons systems, oil/gas/other process industries, and merchant shipping. As fire protection equipment is tested or deployed, emissions of these HFCs occur.

Methodology

A detailed Vintaging Model of ODS-containing equipment and products was used to estimate the actual—versus potential—emissions of various ODS substitutes, including HFCs and PFCs. The name of the model refers to the fact that it tracks the use and emissions of various compounds for the annual “vintages” of new equipment that enter service in each end-use. The Vintaging Model predicts ODS and ODS substitute use in the United States based on modeled estimates of the quantity of equipment or products sold each year containing these chemicals and the amount of the chemical required to manufacture and/or maintain equipment and products over time. Emissions for each end-use were estimated by applying annual leak rates and release profiles, which account for the lag in emissions from equipment as they leak over time. By aggregating the data for 60 different end-uses, the model produces estimates of annual use and emissions of each compound. Further information on the Vintaging Model is contained in Annex 3.9.

Uncertainty and Time-Series Consistency

Given that emissions of ODS substitutes occur from thousands of different kinds of equipment and from millions of point and mobile sources throughout the United States, emission estimates must be made using analytical tools such as the Vintaging Model or the methods outlined in IPCC (2006). Though the model is more comprehensive than the IPCC default methodology, significant uncertainties still exist with regard to the levels of equipment sales, equipment characteristics, and end-use emissions profiles that were used to estimate annual emissions for the various compounds.

The Vintaging Model estimates emissions from 60 end-uses. The uncertainty analysis, however, quantifies the level of uncertainty associated with the aggregate emissions resulting from the top 21 end-uses, comprising over 95 percent of the total emissions, and 6 other end-uses. These 27 end-uses comprise 97 percent of the total emissions, equivalent to 153.3 MMT CO_2 Eq. In an effort to improve the uncertainty analysis, additional end-uses are added annually, with the intention that over time uncertainty for all emissions from the Vintaging Model will be fully characterized. Any end-uses included in previous years’ uncertainty analysis were included in the current uncertainty analysis, whether or not those end-uses were included in the top 95 percent of emissions from ODS Substitutes.

In order to calculate uncertainty, functional forms were developed to simplify some of the complex “vintaging” aspects of some end-use sectors, especially with respect to refrigeration and air-conditioning, and to a lesser degree, fire extinguishing. These sectors calculate emissions based on the entire lifetime of equipment, not just equipment put into commission in the current year, thereby necessitating simplifying equations. The functional forms used variables that included growth rates, emission factors, transition from ODSs, change in charge size as a result of the transition, disposal quantities, disposal emission rates, and either stock for the current year or original ODS consumption. Uncertainty was estimated around each variable within the functional forms based on expert judgment, and a Monte Carlo analysis was performed. The most significant sources of uncertainty for this source category include the emission factors for residential unitary AC, as well as the percent of non-MDI aerosol propellant that is HFC-152a.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-97. Substitution of ozone depleting substances HFC and PFC emissions were estimated to be between 153.0 and 172.3 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 0.22 percent below to 12.4 percent above the emission estimate of 158.6 MMT CO_2 Eq.

Table 4-97: Approach 2 Quantitative Uncertainty Estimates for HFC and PFC Emissions from ODS Substitutes (MMT CO_2 Eq. and Percent)

Source	Gases	2013 Emission Estimate (MMT CO_2 Eq.)[a]	Uncertainty Range Relative to Emission Estimate[b] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Substitution of Ozone Depleting Substances	HFCs and PFCs	158.6	153.0	172.3	-0.22%	+12.4%

[a] 2013 emission estimates and the uncertainty range presented in this table correspond to selected end-uses within the aerosols, foams, solvents, fire extinguishing agents, and refrigerants sectors that comprise 97 percent of total emissions, but not for other remaining categories. Therefore, because the uncertainty associated with emissions from "other" ODS substitutes was not estimated, they were excluded in the uncertainty estimates reported in this table.

[b] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2-equivalent emissions from HFCs and PFCs. The GWPs of N_2O and SF_6 have decreased, leading to a decrease in CO_2-equivalent emissions for these greenhouse gases. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

The decrease in the GWP of HFC-152a and increase in the GWP of all other gases had several impacts on Inventory estimates. In the 1990 through 1991 time period, an overall decrease in total annual GWP-weighted emissions is seen. After 1991, there is an overall increase in total emissions.

In addition, a review of the MVACs, streaming agents, window AC units, ice makers, and small retail food end-uses resulted in revisions to the Vintaging Model since the previous Inventory. Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013.

For the MVAC light-duty vehicle (LDV) and light-duty trucks (LDT) end-uses, operational and servicing leak rates were reduced based on a review of recent literature. For the small retail food and ice makers end-uses, revisions were made to the overall stock, growth rates, assumed transition scenarios, and lifetimes based on research on substitutes and growth in the market. For window air-conditioning, a review of air conditioner sales data from 2002 through 2012 increased the quantity of window air-conditioning equipment introduced into the market for 2002 and 2004 through 2008, while decreasing the quantity of equipment sold into the market for 2003 and 2009 through 2012. In the streaming agents end-use, the assumed transition scenarios were revised based on industry input. Combined, these assumption changes and the use of AR4 GWPs increased GHG emissions on average by 7 percent across the time series.

4.24 Electrical Transmission and Distribution (IPCC Source Category 2G1)

The largest use of sulfur hexafluoride (SF_6), both in the United States and internationally, is as an electrical insulator and interrupter in equipment that transmits and distributes electricity (RAND 2004). The gas has been employed by the electric power industry in the United States since the 1950s because of its dielectric strength and arc-quenching characteristics. It is used in gas-insulated substations, circuit breakers, and other switchgear. SF_6 has replaced flammable insulating oils in many applications and allows for more compact substations in dense urban areas.

Fugitive emissions of SF_6 can escape from gas-insulated substations and switchgear through seals, especially from older equipment. The gas can also be released during equipment manufacturing, installation, servicing, and disposal. Emissions of SF_6 from equipment manufacturing and from electrical transmission and distribution systems were estimated to be 5.1 MMT CO_2 Eq. (0.2 kt) in 2013. This quantity represents an 80 percent decrease from the estimate for 1990 (see Table 4-98 and Table 4-99). There are two potential causes for this decrease: a sharp increase in the price of SF_6 during the 1990s and a growing awareness of the magnitude and environmental impact of SF_6 emissions through programs such as EPA's voluntary SF_6 Emission Reduction Partnership for Electric Power Systems (Partnership) and EPA's GHGRP. Utilities participating in the Partnership have lowered their emission factor (kg SF_6 emitted per kg of nameplate capacity) by more than 75 percent since the Partnership began in 1999. A recent examination of the SF_6 emissions reported by electric power systems to EPA's GHGRP revealed that SF_6 emissions from reporters has decreased by 25 percent from 2011 to 2013, with much of the reduction seen from utilities that are not participants in the Partnership. These utilities may be making relatively large reductions in emissions as they take advantage of relatively large and/or inexpensive emission reduction opportunities (i.e., "low hanging fruit," such as replacing major leaking circuit breakers) that Partners have already taken advantage of under the voluntary program (Ottinger et al. 2014).

Table 4-98: SF_6 Emissions from Electric Power Systems and Electrical Equipment Manufacturers (MMT CO_2 Eq.)

Year	Electric Power Systems	Electrical Equipment Manufacturers	Total
1990	25.1	0.3	25.4
2005	9.8	0.8	10.6
2009	6.7	0.6	7.3
2010	6.2	0.9	7.0
2011	5.7	1.1	6.8
2012	4.6	1.1	5.7
2013	4.2	0.9	5.1

Notes: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
Note: Totals may not sum due to independent rounding.

Table 4-99: SF_6 Emissions from Electric Power Systems and Electrical Equipment Manufacturers (kt)

Year	Emissions
1990	1.1
2005	0.5
2009	0.3
2010	0.3
2011	0.3
2012	0.2

2013	0.2

Methodology

The estimates of emissions from Electrical Transmission and Distribution are comprised of emissions from electric power systems and emissions from the manufacture of electrical equipment. The methodologies for estimating both sets of emissions are described below.

1990 through 1998 Emissions from Electric Power Systems

Emissions from electric power systems from 1990 through 1998 were estimated based on (1) the emissions estimated for this source category in1999, which, as discussed in the next section, were based on the emissions reported during the first year of EPA's SF_6 Emission Reduction Partnership for Electric Power Systems (Partnership), and (2) the RAND survey of global SF_6 emissions. Because most utilities participating in the Partnership reported emissions only for 1999 through 2011, modeling was used to estimate SF_6 emissions from electric power systems for the years 1990 through 1998. To perform this modeling, U.S. emissions were assumed to follow the same trajectory as global emissions from this source during the 1990 to 1999 period. To estimate global emissions, the RAND survey of global SF_6 sales were used, together with the following equation for estimating emissions, which is derived from the mass-balance equation for chemical emissions (Volume 3, Equation 7.3) in the *2006 IPCC Guidelines* (IPCC 2006).[201] (Although Equation 7.3 of the *2006 IPCC Guidelines* appears in the discussion of substitutes for ozone-depleting substances, it is applicable to emissions from any long-lived pressurized equipment that is periodically serviced during its lifetime.)

Emissions (kilograms SF_6) = SF_6 purchased to refill existing equipment (kilograms) + nameplate capacity of retiring equipment (kilograms) [202]

Note that the above equation holds whether the gas from retiring equipment is released or recaptured; if the gas is recaptured, it is used to refill existing equipment, thereby lowering the amount of SF_6 purchased by utilities for this purpose.

Gas purchases by utilities and equipment manufacturers from 1961 through 2003 are available from the RAND (2004) survey. To estimate the quantity of SF_6 released or recovered from retiring equipment, the nameplate capacity of retiring equipment in a given year was assumed to equal 81.2 percent of the amount of gas purchased by electrical equipment manufacturers 40 years previous (e.g., in 2000, the nameplate capacity of retiring equipment was assumed to equal 81.2 percent of the gas purchased in 1960). The remaining 18.8 percent was assumed to have been emitted at the time of manufacture. The 18.8 percent emission factor is an average of IPCC default SF_6 emission rates for Europe and Japan for 1995 (IPCC 2006). The 40-year lifetime for electrical equipment is also based on IPCC (2006). The results of the two components of the above equation were then summed to yield estimates of global SF_6 emissions from 1990 through 1999.

U.S. emissions between 1990 and 1999 are assumed to follow the same trajectory as global emissions during this period. To estimate U.S. emissions, global emissions for each year from 1990 through 1998 were divided by the estimated global emissions from 1999. The result was a time series of factors that express each year's global emissions as a multiple of 1999 global emissions. Historical U.S. emissions were estimated by multiplying the factor for each respective year by the estimated U.S. emissions of SF_6 from electric power systems in 1999 (estimated to be 14.3 MMT CO_2 Eq.).

Two factors may affect the relationship between the RAND sales trends and actual global emission trends. One is utilities' inventories of SF_6 in storage containers. When SF_6 prices rise, utilities are likely to deplete internal inventories before purchasing new SF_6 at the higher price, in which case SF_6 sales will fall more quickly than emissions. On the other hand, when SF_6 prices fall, utilities are likely to purchase more SF_6 to rebuild inventories, in which case sales will rise more quickly than emissions. This effect was accounted for by applying 3-year

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[201] Ideally, sales to utilities in the U.S. between 1990 and 1999 would be used as a model. However, this information was not available. There were only two U.S. manufacturers of SF_6 during this time period, so it would not have been possible to conceal sensitive sales information by aggregation.

[202] Nameplate capacity is defined as the amount of SF_6 within fully charged electrical equipment.

smoothing to utility SF_6 sales data. The other factor that may affect the relationship between the RAND sales trends and actual global emissions is the level of imports from and exports to Russia and China. SF_6 production in these countries is not included in the RAND survey and is not accounted for in any another manner by RAND. However, atmospheric studies confirm that the downward trend in estimated global emissions between 1995 and 1998 was real (see the Uncertainty discussion below).

1999 through 2013 Emissions from Electric Power Systems

Emissions from electric power systems from 1999 to 2013 were estimated based on: (1) reporting from utilities participating in EPA's SF_6 Emission Reduction Partnership for Electric Power Systems (Partners), which began in 1999; (2) reporting from utilities covered by the EPA's GHGRP, which began in 2012 for emissions occurring in 2011 (GHGRP-Only Reporters); and (3) the relationship between utilities' reported emissions and their transmission miles as reported in the 2001, 2004, 2007, 2010, and 2013 Utility Data Institute (UDI) Directories of Electric Power Producers and Distributors (UDI 2001, 2004, 2007, 2010, 2013), which was applied to the electric power systems that do not report to EPA (Non-Reporters). (Transmission miles are defined as the miles of lines carrying voltages above 34.5 kV).

Partners

Over the period from 1999 to 2013, Partner utilities, which for inventory purposes are defined as utilities that either currently are or previously have been part of the Partnership, represented between 42 percent and 48 percent of total U.S. transmission miles. Partner utilities estimated their emissions using a Tier 3 utility-level mass balance approach (IPCC 2006). If a Partner utility did not provide data for a particular year, emissions were interpolated between years for which data were available or extrapolated based on Partner-specific transmission mile growth rates. In 2012, many Partners began reporting their emissions (for 2011 and later years) through EPA's GHGRP (discussed further below) rather than through the Partnership. In 2013, approximately 0.3 percent of the total emissions attributed to Partner utilities were reported through Partnership reports. Approximately 91 percent of the total emissions attributed to Partner utilities were reported and verified through EPA's GHGRP. Partners without verified 2013 data accounted for approximately 9 percent of the total emissions attributed to Partner utilities.[203]

GHGRP-Only Reporters

EPA's GHGRP requires users of SF_6 in electric power systems to report emissions if the facility has a total SF_6 nameplate capacity that exceeds 17,820 pounds. (This quantity is the nameplate capacity that would result in annual SF_6 emissions equal to 25,000 metric tons of CO_2 equivalent at the historical emission rate reported under the Partnership.) As under the Partnership, electric power systems that report their SF_6 emissions under EPA's GHGRP are required to use the Tier 3 utility-level mass-balance approach. Many Partners began reporting their emissions through EPA's GHGRP in 2012 (reporting emissions for 2011 and later years) because their nameplate capacity exceeded the reporting threshold. Partners who did not report through EPA's GHGRP continued to report through the Partnership.

In addition, many non-Partners began reporting to EPA for the first time through its GHGRP in 2012. Non-Partner emissions reported and verified under EPA's GHGRP were compiled to form a new category of reported data

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[203] It should be noted that data reported through the GHGRP must go through a verification process; only data verified as of September 1, 2014 could be used in the emission estimates for 2013. For Partners whose GHGRP data was not yet verified, emissions were extrapolated based upon historical Partner-specific transmission mile growth rates, and those Partners are included in the 'non-reporting Partners' category.

For electric power systems, verification involved a series of electronic range, completeness, and algorithm checks for each report submitted. In addition, EPA manually reviewed the reported data and compared each facility's reported transmission miles with the corresponding quantity in the UDI 2013 database (UDI 2013). In the first year of GHGRP reporting, EPA followed up with reporters where the discrepancy between the reported miles and the miles published by UDI was greater than 10 percent, with a goal to improve data quality. Only GHGRP data verified as of September 1, 2014 was included in the emission estimates for 2011, 2012, and 2013.

(GHGRP-Only Reporters). GHGRP-Only Reporters accounted for 24 percent of U.S. transmission miles and 26 percent of estimated U.S. emissions from electric power system in 2013.[204]

Non-Reporters

Emissions from Non-Reporters (i.e., utilities other than Partners and GHGRP-Only Reporters) in every year since 1999 were estimated using the results of a regression analysis that correlated emissions from reporting utilities (using verified data from both Partners and GHGRP-Only Reporters) with their transmission miles.[205] Two equations were developed, one for "non-large" and one for "large" utilities (i.e., with fewer or greater than 10,000 transmission miles, respectively). The distinction between utility sizes was made because the regression analysis showed that the relationship between emissions and transmission miles differed for non-large and large transmission networks. As noted above, non-Partner emissions were reported to the EPA for the first time through its GHGRP in 2012 (representing 2011 emissions). This set of reported data was of particular interest because it provided insight into the emission rate of non-Partners, which previously was assumed to be equal to the historical (1999) emission rate of Partners for both large and non-large utilities.[206] The availability of non-Partner emissions estimates allowed the regression analysis to be modified for both large and non-large groups. Specifically, emissions were estimated for Non-Reporters as follows:

- Non-Reporters, 1999 to 2011: First, the 2011 emission rates (per kg nameplate capacity and per transmission mile) reported by Partners and GHGRP-Only Reporters were reviewed to determine whether there was a statistically significant difference between these two groups. Transmission mileage data for 2011 was reported through GHGRP, with the exception of transmission mileage data for Partners that did not report through GHGRP, which was obtained from UDI. It was determined that there is no statistically significant difference between the emission rates of Partners and GHGRP-Only reporters; therefore, Partner and GHGRP-Only reported data for 2011 were combined to develop regression equations to estimate the emissions of Non-Reporters for both "non-large" and "large" utilities. Historical emissions from Non-Reporters for both "non-large" and "large" utilities were estimated by linearly interpolating between the 1999 regression coefficients (based on 1999 Partner data) and the 2011 regression coefficients.

- Non-Reporters, 2012 - Present: It was determined that there continued to be no statistically significant difference between the emission rates reported by Partners and by GHGRP-Only Reporters. Therefore, the emissions data from both groups were combined to develop regression equations for 2012. This was repeated for 2013 using Partner and GHGRP-Only Reporter data for 2013.

    - *"Non-large" utilities (less than 10,000 transmission miles):* The 2013 regression equation for "non-large" utilities was developed based on the emissions reported by a subset of 89 Partner utilities and GHGRP-Only utilities (representing approximately 47 percent of total U.S. transmission miles for utilities with fewer than 10,000 transmission miles). The regression equation for 2013 is:

$$\text{Emissions (kg)} = 0.217 \times \text{Transmission Miles}$$

    - *"Large" utilities (more than 10,000 transmission miles)*: The 2013 regression equation was developed based on the emissions reported by a subset of 17 Partners and GHGRP-only utilities (representing approximately 83 percent of total U.S. transmission miles for utilities with greater than 10,000 transmission miles). The regression equation for 2013 is:

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[204] Also, GHGRP-reported emissions from 17 facilities that had one or fewer transmission miles were included in the emission estimates for 2011. Emissions from these facilities comprise approximately 1.2 percent of total reported and verified emissions. In 2012, 16 facilities had one or fewer transmission miles, comprising 1.4 percent of verified emissions and in 2013, 16 facilities had one or fewer transmission miles, comprising 3.2 percent of verified emissions. These facilities were not included in the development of the regression equations (discussed further below). EPA is continuing to investigate whether or not these emissions are already implicitly accounted for in the relationship between transmission miles and emissions, and whether to update the regression analysis to better capture emissions from non-reporters that may have zero transmission miles.

[205] In the United States, SF_6 is contained primarily in transmission equipment rated above 34.5 kV.

[206] Partners in EPA's SF_6 Emission Reduction Partnership reduced their emissions by approximately 77 percent from 1999 to 2013.

$$\text{Emissions (kg)} = 0.225 \times \text{Transmission Miles}$$

Table 4-4-100 below shows the percentage of transmission miles covered by reporters (i.e., associated with reported data) and the regression coefficient for both large and non-large reporters for 1999 (the first year data was reported), and for 2011 through 2013 (the first three years with GHGRP reported data). The coefficients for non-large utilities and large utilities both decreased slightly between 2012 and 2013.

Table 4-4-100: Transmission Mile Coverage and Regression Coefficients for Large and Non-Large Utilities, Percent

	Non-large				Large			
	1999	2011	2012	2013	1999	2011	2012	2013
Percentage of Miles Covered by Reporters	31	45	44	47	86	97	88	83
Regression Coefficient[a]	0.89	0.33	0.23	0.22	0.58	0.27	0.24	0.22

[a] Regression coefficient is defined as emissions (in kg) divided by transmission miles.

Note: "Non-large" represents reporters with fewer than 10,000 transmission miles.

Data on transmission miles for each Non-Reporter for the years 2000, 2003, 2006, and 2009, and 2012 were obtained from the 2001, 2004, 2007, 2010, and 2013 UDI Directories of Electric Power Producers and Distributors, respectively (UDI 2001, 2004, 2007, 2010, 2013). The U.S. transmission system grew by over 25,000 miles between 2000 and 2003 yet declined by almost 4,000 miles between 2003 and 2006. Given these fluctuations, periodic increases are assumed to occur gradually. Therefore, transmission mileage was assumed to increase at an annual rate of 1.2 percent between 2000 and 2003 and decrease by -0.20 percent between 2003 and 2006. This growth rate grew to 3 percent from 2006 to 2009 as transmission miles increased by more than 59,000 miles. The annual growth rate for 2009 through 2012 was calculated to be 2.0 percent as transmission miles grew by approximately 43,000 during this time period.

Total Industry Emissions

As a final step, total electric power system emissions from 1999 through 2013 were determined for each year by summing the Partner reported and estimated emissions (reported data was available through the EPA's SF_6 Emission Reduction Partnership for Electric Power Systems), the GHGRP-Only reported emissions, and the non-reporting utilities' emissions (determined using the regression equations).

1990 through 2013 Emissions from Manufacture of Electrical Equipment

The 1990 to 2013 emission estimates for original equipment manufacturers (OEMs) were derived by assuming that manufacturing emissions equal 10 percent of the quantity of SF_6 provided with new equipment. The quantity of SF_6 provided with new equipment was estimated based on statistics compiled by the National Electrical Manufacturers Association (NEMA). These statistics were provided for 1990 to 2000; the quantities of SF_6 provided with new equipment for 2001 to 2013 were estimated using Partner reported data and the total industry SF_6 nameplate capacity estimate (198.2 MMT CO_2 Eq. in 2013). Specifically, the ratio of new nameplate capacity to total nameplate capacity of a subset of Partners for which new nameplate capacity data was available from 1999 to 2013 was calculated. These ratios were then multiplied by the total industry nameplate capacity estimate for each year to derive the amount of SF_6 provided with new equipment for the entire industry. The 10 percent emission rate is the average of the "ideal" and "realistic" manufacturing emission rates (4 percent and 17 percent, respectively) identified in a paper prepared under the auspices of the International Council on Large Electric Systems (CIGRE) in February 2002 (O'Connell et al. 2002).

Uncertainty and Time-Series Consistency

To estimate the uncertainty associated with emissions of SF_6 from Electrical Transmission and Distribution, uncertainties associated with four quantities were estimated: (1) emissions from Partners, (2) emissions from GHGRP-Only Reporters, (3) emissions from Non-Reporters, and (4) emissions from manufacturers of electrical equipment. A Monte Carlo analysis was then applied to estimate the overall uncertainty of the emissions estimate.

Total emissions from the SF_6 Emission Reduction Partnership include emissions from both reporting (through the Partnership or GHGRP) and non-reporting Partners. For reporting Partners, individual Partner-reported SF_6 data was assumed to have an uncertainty of 10 percent. Based on a Monte Carlo analysis, the cumulative uncertainty of all Partner-reported data was estimated to be 2.5 percent. The uncertainty associated with extrapolated or interpolated emissions from non-reporting Partners was assumed to be 20 percent.

For GHGRP-Only Reporters, reported SF_6 data was assumed to have an uncertainty of 20 percent.[207] Based on a Monte Carlo analysis, the cumulative uncertainty of all GHGRP-Only reported data was estimated to be 5.8 percent.

There are two sources of uncertainty associated with the regression equations used to estimate emissions in 2013 from Non-Reporters: (1) uncertainty in the coefficients (as defined by the regression standard error estimate), and (2) the uncertainty in total transmission miles for Non-Reporters. Uncertainties were also estimated regarding (1) the quantity of SF_6 supplied with equipment by equipment manufacturers, which is projected from Partner provided nameplate capacity data and industry SF_6 nameplate capacity estimates, and (2) the manufacturers' SF_6 emissions rate.

The results of the Approach 2 quantitative uncertainty analysis are summarized in Table 4-101. Electrical Transmission and Distribution SF_6 emissions were estimated to be between 4.0 and 6.0 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 20 percent below and 19 percent above the emission estimate of 5.1 MMT CO_2 Eq.

Table 4-101: Approach 2 Quantitative Uncertainty Estimates for SF_6 Emissions from Electrical Transmission and Distribution (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to 2013 Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
Electrical Transmission and Distribution	SF_6	5.1	4.0	6.0	-20%	+19%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

In addition to the uncertainty quantified above, there is uncertainty associated with using global SF_6 sales data to estimate U.S. emission trends from 1990 through 1999. However, the trend in global emissions implied by sales of SF_6 appears to reflect the trend in global emissions implied by changing SF_6 concentrations in the atmosphere. That is, emissions based on global sales declined by 29 percent between 1995 and 1998 (RAND 2004), and emissions based on atmospheric measurements declined by 17 percent over the same period (Levin et al. 2010).

Several pieces of evidence indicate that U.S. SF_6 emissions were reduced as global emissions were reduced. First, the decreases in sales and emissions coincided with a sharp increase in the price of SF_6 that occurred in the mid-1990s and that affected the United States as well as the rest of the world. A representative from DILO, a major manufacturer of SF_6 recycling equipment, stated that most U.S. utilities began recycling rather than venting SF_6 within two years of the price rise. Finally, the emissions reported by the one U.S. utility that reported its emissions for all the years from 1990 through 1999 under the Partnership showed a downward trend beginning in the mid-1990s.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.

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[207] Uncertainty is assumed to be higher for the GHGRP-Only category, because 2011 is the first year that those utilities have reported to EPA.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2 Eq. emissions from CH_4, HFCs, and PFCs. The GWPs of N_2O and SF_6 have decreased, leading to a decrease in CO_2 Eq. emissions for SF_6. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

Only taking this change into consideration, emissions estimates for each year from 1990 to 2012 would have slightly decreased, relative to the emissions estimates in the previous Inventory report. However, other changes to the historical calculations, as noted below, resulted in emission estimates fluctuating slightly (increasing for some years and decreasing for other years) across the time series.

The historical emissions estimated for this source category have undergone several minor revisions. SF_6 emission estimates for the period 1990 through 2012 were updated relative to the previous report based on revisions to interpolated and extrapolated non-reported Partner data as well as resubmissions of estimates through the GHGRP for 2011 and 2012.[208] The previously-described interpolation between 1999 and 2012 regression coefficients to estimate emissions from non-reporting utilities were updated using revised GHGRP reports, which impacted historical estimates for the period 2000 through 2012. Additionally, updated leak rates were calculated from resubmitted Partner data through the GHGRP. These leak rates are used to estimate the nameplate capacity of non-reporters during these years, and are interpolated back through 1999 to calculate Non-Reporter nameplate capacity over the entire time series.[209] Finally, revisions were made regarding the incorporation of transmission mile data from the UDI database to remove instances of double counting transmission miles between parent and subsidiary companies. Reductions in the total transmission miles reduced the total number of non-reporter transmission miles, which reduced non-reporter emissions, and therefore total emissions.

As a result of the recalculations, SF_6 emissions from electrical transmission and distribution decreased by 6 percent for 2012 relative to the previous report. On average, the change in SF_6 emission estimates for the entire time series is approximately 0.5 percent per year.

Planned Improvements

EPA is exploring the use of OEM data that is reported under EPA's GHGRP to use for future Inventory reports instead of estimating those emissions based on elements reported by utilities to the GHGRP and Partner data. Specifically, using the GHGRP-reported OEM emissions and the estimated nameplate capacity increase estimated for users of electrical equipment (available in the existing methodology), a leak rate would be calculated. This approach would require estimating the portion of industry not reporting to the GHGRP program, which would require market research. Once a new leak rate is established, leak rates could be interpolated for years between 2000 (at 10 percent) and 2011. In implementing improvements and integration of data from EPA's GHGRP, the latest guidance from the IPCC on the use of facility-level data in national inventories will be relied upon.[210]

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[208] The earlier year estimates within the time series (i.e., 1990-1998) were updated based on revisions to the 1999 U.S. emission estimate because emissions for 1990-1998 are estimated by multiplying a series of annual factors by the estimated U.S. emissions of SF_6 from electric power systems in 1999 (see Methodology section).

[209] Nameplate capacity estimates affect sector emissions because OEM emission estimation is calculated using total industry nameplate capacity.

[210] See <http://www.ipcc-nggip.iges.or.jp/meeting/pdfiles/1008_Model_and_Facility_Level_Data_Report.pdf>.

Box 4-2: Potential Emission Estimates of HFCs, PFCs, SF_6, and NF_3

Emissions of HFCs, PFCs, SF_6, and NF_3 from industrial processes can be estimated in two ways, either as potential emissions or as actual emissions. Emission estimates in this chapter are "actual emissions," which are defined by the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006) as estimates that take into account the time lag between consumption and emissions. In contrast, "potential emissions" are defined to be equal to the amount of a chemical consumed in a country, minus the amount of a chemical recovered for destruction or export in the year of consideration. Potential emissions will generally be greater for a given year than actual emissions, since some amount of chemical consumed will be stored in products or equipment and will not be emitted to the atmosphere until a later date, if ever. Although actual emissions are considered to be the more accurate estimation approach for a single year, estimates of potential emissions are provided for informational purposes.

Separate estimates of potential emissions were not made for industrial processes that fall into the following categories:

- *By-product emissions*. Some emissions do not result from the consumption or use of a chemical, but are the unintended by-products of a process. For such emissions, which include emissions of CF_4 and C_2F_6 from aluminum production and of HFC-23 from HCFC-22 production, the distinction between potential and actual emissions is not relevant.
- *Potential emissions that equal actual emissions*. For some sources, such as magnesium production and processing, no delay between consumption and emission is assumed and, consequently, no destruction of the chemical takes place. In this case, actual emissions equal potential emissions.

Table 4-102 presents potential emission estimates for HFCs and PFCs from the substitution of ozone depleting substances, HFCs, PFCs, SF_6, and NF_3 from semiconductor manufacture, and SF_6 from magnesium production and processing and electrical transmission and distribution.[211] Potential emissions associated with the substitution for ozone depleting substances were calculated using the EPA's Vintaging Model. Estimates of HFCs, PFCs, and SF_6 consumed by semiconductor manufacture were developed by dividing chemical-by-chemical emissions by the appropriate chemical-specific emission factors from the *2006 IPCC Guidelines* (Tier 2c). Estimates of CF_4 consumption were adjusted to account for the conversion of other chemicals into CF_4 during the semiconductor manufacturing process, again using the default factors from the *2006 IPCC Guidelines*. Potential SF_6 emissions estimates for electrical transmission and distribution were developed using U.S. utility purchases of SF_6 for electrical equipment. From 1999 through 2013, estimates were obtained from reports submitted by participants in EPA's SF_6 Emission Reduction Partnership for Electric Power Systems as well as EPA's Greenhouse Gas Reporting Program (GHGRP). U.S. utility purchases of SF_6 for electrical equipment from 1990 through 1998 were backcasted based on world sales of SF_6 to utilities. Purchases of SF_6 by utilities were added to SF_6 purchases by electrical equipment manufacturers to obtain total SF_6 purchases by the electrical equipment sector.

Table 4-102: 2013 Potential and Actual Emissions of HFCs, PFCs, SF_6, and NF_3 from Selected Sources (MMT CO_2 Eq.)

Source	Potential	Actual
Substitution of Ozone Depleting Substances	306.9	158.6
Aluminum Production	NA	3.0
HCFC-22 Production	NA	4.1
Semiconductor Manufacture	43.7	4.0
Magnesium Production and Processing	1.5	1.5
Electrical Transmission and Distribution	33.3	5.1

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.
NA - Not applicable.

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[211] See Annex 5 for a discussion of sources of SF_6 emissions excluded from the actual emissions estimates in this report.

Under EPA's GHGRP, producers and larger importers and exporters[212] of fluorinated greenhouse gases (F-GHG) in bulk began annually reporting their production, destruction, imports, and exports in 2011 (for 2010 supplies), and larger importers and exporters of F-GHGs inside of pre-charged equipment began reporting their imports and exports in 2012 (for 2011 supplies). The collection of data from both emitters and suppliers of F-GHGs enables the comparison of consumption that is implied by emissions (downstream estimation method) to the consumption that is implied by balancing of production, destruction, imports, and exports (upstream estimation method). This type of comparison ultimately supports and improves estimates of emissions, as noted in the *2006 IPCC Guidelines*:

> "[W]hen considered along with estimates of actual emissions, the potential emissions approach can assist in validation of completeness of sources covered and as a QC check by comparing total domestic consumption as calculated in this 'potential emissions approach' per compound with the sum of all activity data of the various uses (IPCC 2006)."

A comparison of upstream and downstream consumption estimates of SF_6 was performed to help evaluate the accuracy and completeness of the emissions inventory. This analysis revealed that the two potential emissions estimates for 2012 (the upstream estimation and downstream estimation methods) differed with the supply-based, upstream consumption estimate significantly larger than emitter-based, downstream consumption estimate (Ottinger et al. 2014). This finding indicates that methods for determining national SF_6 actual emission estimates by industry sector are generating results that, when summed, do not fall within a close proximity to the overall total U.S. supply of SF_6 gas.

While multiple sources of uncertainty affect both data sets, Ottinger et al (2014) conclude that current SF_6 emission estimates likely do not account for all significant sources of SF_6 in the United States. Additional research is necessary to identify the other significant applications that consume and emit SF_6.

4.25 Nitrous Oxide from Product Uses (IPCC Source Category 2G3)

N_2O is a clear, colorless, oxidizing liquefied gas, with a slightly sweet odor which is used in a wide variety of specialized product uses and applications. The amount of N_2O that is actually emitted depends upon the specific product use or application.

There are a total of three N_2O production facilities currently operating in the United States (Ottinger 2014). N_2O is primarily used in carrier gases with oxygen to administer more potent inhalation anesthetics for general anesthesia, and as an anesthetic in various dental and veterinary applications. The second main use of N_2O is as a propellant in pressure and aerosol products, the largest application being pressure-packaged whipped cream. Small quantities of N_2O also are used in the following applications:

- Oxidizing agent and etchant used in semiconductor manufacturing;
- Oxidizing agent used, with acetylene, in atomic absorption spectrometry;
- Production of sodium azide, which is used to inflate airbags;
- Fuel oxidant in auto racing; and
- Oxidizing agent in blowtorches used by jewelers and others (Heydorn 1997).

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[212] Importers and exporters report only if either their total imports or their total exports of F-GHGs are greater than or equal to 25,000 metric tons of CO_2 Eq. per year

Production of N_2O in 2013 was approximately 15 kt (Table 4-103).

Table 4-103: N_2O Production (kt)

Year	kt
1990	16
2005	15
2009	15
2010	15
2011	15
2012	15
2013	15

N_2O emissions were 4.2 MMT CO_2 Eq. (14 kt) in 2013 (Table 4-104). Production of N_2O stabilized during the 1990s because medical markets had found other substitutes for anesthetics, and more medical procedures were being performed on an outpatient basis using local anesthetics that do not require N_2O. The use of N_2O as a propellant for whipped cream has also stabilized due to the increased popularity of cream products packaged in reusable plastic tubs (Heydorn 1997).

Table 4-104: N_2O Emissions from N_2O Product Usage (MMT CO_2 Eq. and kt)

Year	MMT CO_2 Eq.	kt
1990	4.2	14
2005	4.2	14
2009	4.2	14
2010	4.2	14
2011	4.2	14
2012	4.2	14
2013	4.2	14

Note: Emissions values are presented in CO_2 equivalent mass units using IPCC AR4 GWP values.

Methodology

Emissions from N_2O product uses were estimated using the following equation:

$$E_{pu} = \sum_{a} (P \times S_a \times ER_a)$$

where,

E_{pu} = N_2O emissions from product uses, metric tons
P = Total U.S. production of N_2O, metric tons
a = specific application
S_a = Share of N_2O usage by application *a*
ER_a = Emission rate for application *a*, percent

The share of total quantity of N_2O usage by end use represents the share of national N_2O produced that is used by the specific subcategory (i.e., anesthesia, food processing, etc.). In 2013, the medical/dental industry used an estimated 86.5 percent of total N_2O produced, followed by food processing propellants at 6.5 percent. All other categories combined used the remainder of the N_2O produced. This subcategory breakdown has changed only slightly over the past decade. For instance, the small share of N_2O usage in the production of sodium azide has declined significantly during the 1990s. Due to the lack of information on the specific time period of the phase-out in this market subcategory, most of the N_2O usage for sodium azide production is assumed to have ceased after 1996, with the majority of its small share of the market assigned to the larger medical/dental consumption subcategory (Heydorn 1997). The N_2O was allocated across the following categories: medical applications, food processing propellant, and sodium azide production (pre-1996). A usage emissions rate was then applied for each sector to estimate the amount of N_2O emitted.

Only the medical/dental and food propellant subcategories were estimated to release emissions into the atmosphere, and therefore these subcategories were the only usage subcategories with emission rates. For the medical/dental subcategory, due to the poor solubility of N_2O in blood and other tissues, none of the N_2O is assumed to be metabolized during anesthesia and quickly leaves the body in exhaled breath. Therefore, an emission factor of 100 percent was used for this subcategory (IPCC 2006). For N_2O used as a propellant in pressurized and aerosol food products, none of the N_2O is reacted during the process and all of the N_2O is emitted to the atmosphere, resulting in an emission factor of 100 percent for this subcategory (IPCC 2006). For the remaining subcategories, all of the N_2O is consumed/reacted during the process, and therefore the emission rate was considered to be zero percent (Tupman 2002).

The 1990 through 1992 N_2O production data were obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997). N_2O production data for 1993 through 1995 were not available. Production data for 1996 was specified as a range in two data sources (Heydorn 1997, Tupman 2002). In particular, for 1996, Heydorn (1997) estimates N_2O production to range between 13.6 and 18.1 thousand metric tons. Tupman (2003) provided a narrower range (15.9 to 18.1 thousand metric tons) for 1996 that falls within the production bounds described by Heydorn (1997). Tupman (2003) data are considered more industry-specific and current. Therefore, the midpoint of the narrower production range was used to estimate N_2O emissions for years 1993 through 2001 (Tupman 2003). The 2002 and 2003 N_2O production data were obtained from the Compressed Gas Association Nitrous Oxide Fact Sheet and Nitrous Oxide Abuse Hotline (CGA 2002, 2003). These data were also provided as a range. For example, in 2003, CGA (2003) estimates N_2O production to range between 13.6 and 15.9 thousand metric tons. Due to the unavailability of data, production estimates for years 2004 through 2013 were held constant at the 2003 value.

The 1996 share of the total quantity of N_2O used by each subcategory was obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997). The 1990 through 1995 share of total quantity of N_2O used by each subcategory was kept the same as the 1996 number provided by SRI Consulting. The 1997 through 2001share of total quantity of N_2O usage by sector was obtained from communication with a N_2O industry expert (Tupman 2002). The 2002 and 2003 share of total quantity of N_2O usage by sector was obtained from CGA (2002, 2003). Due to the unavailability of data, the share of total quantity of N_2O usage data for years 2004 through 2013 was assumed to equal the 2003 value. The emissions rate for the food processing propellant industry was obtained from SRI Consulting's Nitrous Oxide, North America report (Heydorn 1997), and confirmed by a N_2O industry expert (Tupman 2002). The emissions rate for all other subcategories was obtained from communication with a N_2O industry expert (Tupman 2002). The emissions rate for the medical/dental subcategory was obtained from the *2006 IPCC Guidelines*.

Uncertainty and Time-Series Consistency

The overall uncertainty associated with the 2013 N_2O emission estimate from N_2O product usage was calculated using the *2006 IPCC Guidelines* (2006) Approach 2 methodology. Uncertainty associated with the parameters used to estimate N_2O emissions include production data, total market share of each end use, and the emission factors applied to each end use, respectively.

The results of this Approach 2 quantitative uncertainty analysis are summarized in Table 4-105. N_2O emissions from N_2O product usage were estimated to be between 3.2 and 5.2 MMT CO_2 Eq. at the 95 percent confidence level. This indicates a range of approximately 24 percent below to 24 percent above the emission estimate of 4.2 MMT CO_2 Eq.

Table 4-105: Approach 2 Quantitative Uncertainty Estimates for N_2O Emissions from N_2O Product Usage (MMT CO_2 Eq. and Percent)

Source	Gas	2013 Emission Estimate (MMT CO_2 Eq.)	Uncertainty Range Relative to Emission Estimate[a] (MMT CO_2 Eq.)		(%)	
			Lower Bound	Upper Bound	Lower Bound	Upper Bound
N_2O Product Use	N_2O	4.2	3.2	5.2	-24%	+24%

[a] Range of emission estimates predicted by Monte Carlo Stochastic Simulation for a 95 percent confidence interval.

Furthermore, methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time-series are described in more detail in the Methodology section, above.

Recalculations Discussion

For the current Inventory, emission estimates have been revised to reflect the GWPs provided in the *IPCC Fourth Assessment Report* (AR4) (IPCC 2007). AR4 GWP values differ slightly from those presented in the *IPCC Second Assessment Report* (SAR) (IPCC 1996) (used in the previous inventories) which results in time-series recalculations for most inventory sources. Under the most recent reporting guidelines (UNFCCC 2014), countries are required to report using the AR4 GWPs, which reflect an updated understanding of the atmospheric properties of each greenhouse gas. The GWPs of CH_4 and most fluorinated greenhouse gases have increased, leading to an overall increase in CO_2-equivalent emissions from CH_4, HFCs, and PFCs. The GWPs of N_2O and SF_6 have decreased, leading to a decrease in CO_2-equivalent emissions for N_2O. The AR4 GWPs have been applied across the entire time series for consistency. For more information please see the Recalculations and Improvements Chapter.

Planned Improvements

Planned improvements include a continued evaluation of alternative production statistics for cross verification, a reassessment of N_2O product use subcategories to accurately represent trends, investigation of production and use cycles, and the potential need to incorporate a time lag between production and ultimate product use and resulting release of N_2O. Additionally, planned improvements include considering imports and exports of N_2O for product uses.

Future Inventories will examine data from EPA's GHGRP to improve the emission estimates for the N_2O product use subcategory. Particular attention will be made to ensure time series consistency, as the facility-level reporting data from EPA's GHGRP are not available for all inventory years as reported in this Inventory.

4.26 Industrial Processes and Product Use Sources of Indirect Greenhouse Gases

In addition to the main greenhouse gases addressed above, many industrial processes can result in emissions of various ozone precursors (i.e., indirect greenhouse gases). As some of industrial applications also employ thermal incineration as a control technology, combustion by-products, such as carbon monoxide (CO) and nitrogen oxides (NO_x), are also reported with this source category. Non-CH_4 volatile organic compounds (NMVOCs), commonly referred to as "hydrocarbons," are the primary gases emitted from most processes employing organic or petroleum based products, and can also result from the product storage and handling. Accidental releases of greenhouse gases associated with product use and handling can constitute major emissions in this category. In the United States, emissions from product use are primarily the result of solvent evaporation, whereby the lighter hydrocarbon molecules in the solvents escape into the atmosphere. The major categories of product uses include: degreasing, graphic arts, surface coating, other industrial uses of solvents (e.g., electronics), dry cleaning, and non-industrial uses (e.g., uses of paint thinner). Product usage in the United States also results in the emission of small amounts of hydrofluorocarbons (HFCs) and hydrofluoroethers (HFEs), which are included under Substitution of Ozone Depleting Substances in this chapter.

Total emissions of nitrogen oxides (NO$_x$), carbon monoxide (CO), and non-CH$_4$ volatile organic compounds (NMVOCs) from non-energy industrial processes and product use from 1990 to 2013 are reported in Table 4-106.

Table 4-106: NO$_x$, CO, and NMVOC Emissions from Industrial Processes and Product Use (kt)

Gas/Source	1990	2005	2009	2010	2011	2012	2013
NO$_x$	**653**	**631**	**544**	**521**	**498**	**498**	**498**
Industrial Processes							
Other Industrial Processes	378	482	395	374	353	353	353
Metals Processing	97	66	76	73	71	71	71
Chemical and Allied Product Manufacturing	168	61	54	53	51	51	51
Storage and Transport	3	16	13	16	20	20	20
Miscellaneous[a]	6	2	2	2	3	3	3
Product Use							
Surface Coating	1	3	3	2	1	1	1
Graphic Arts	+	0	0	0	0	0	0
Degreasing	+	0	0	0	0	0	0
Dry Cleaning	+	0	0	0	0	0	0
Other Industrial Processes[b]	+	0	0	0	0	0	0
Non-Industrial Processes[c]	+	0	0	0	0	0	0
Other	NA	0	0	0	0	0	0
CO	**4,552**	**1,716**	**1,467**	**1,411**	**1,355**	**1,355**	**1,355**
Industrial Processes							
Metals Processing	2,640	829	815	791	766	766	766
Other Industrial Processes	537	534	397	367	337	337	337
Chemical and Allied Product Manufacturing	1,183	208	178	173	167	167	167
Miscellaneous[a]	111	36	51	53	56	56	56
Storage and Transport	76	107	21	24	27	27	27
Product Use							
Surface Coating	1	2	5	3	2	2	2
Other Industrial Processes[b]	4	0	0	0	0	0	0
Dry Cleaning	+	0	0	0	0	0	0
Degreasing	+	0	0	0	0	0	0
Graphic Arts	+	0	0	0	0	0	0
Non-Industrial Processes[c]	+	0	0	0	0	0	0
Other	NA	0	0	0	0	0	0
NMVOCs	**8,419**	**6,448**	**4,781**	**4,556**	**4,331**	**4,331**	**4,331**
Industrial Processes							
Storage and Transport	1,490	1,442	1,143	1,093	1,043	1,043	1,043
Other Industrial Processes	401	457	351	340	329	329	329
Chemical and Allied Product Manufacturing	634	235	86	85	83	83	83
Metals Processing	122	49	36	35	34	34	34
Miscellaneous[a]	22	19	28	29	30	30	30
Product Use							
Surface Coating	2,523	1,739	1,285	1,218	1,152	1,152	1,152
Non-Industrial Processes[c]	1,900	1,594	1,177	1,116	1,055	1,055	1,055
Degreasing	744	309	228	217	205	205	205
Dry Cleaning	215	254	187	178	168	168	168

Graphic Arts	274	213	158	149	141	141	141
Other Industrial Processes[b]	94	97	71	68	64	64	64
Other	0	39	29	28	26	26	26

[a] Miscellaneous includes the following categories: catastrophic/accidental release, other combustion, health services, cooling towers, and fugitive dust. It does not include agricultural fires or slash/prescribed burning, which are accounted for under the Field Burning of Agricultural Residues source.

[b] Includes rubber and plastics manufacturing, and other miscellaneous applications.

[c] Includes cutback asphalt, pesticide application adhesives, consumer solvents, and other miscellaneous applications.

+ Does not exceed 0.05 MMT CO_2 Eq.

Note: Totals may not sum due to independent rounding.

Methodology

Emission estimates for 1990 through 2013 were obtained from data published on the National Emission Inventory (NEI) Air Pollutant Emission Trends web site (EPA 2015), and disaggregated based on EPA (2003). Data were collected for emissions of carbon monoxide (CO), nitrogen oxides (NOx), volatile organic compounds (VOC), and sulfur dioxide (SO_2) from metals processing, chemical manufacturing, other industrial processes, transport and storage, and miscellaneous sources. Emission estimates for 2013 for non-EGU and non-mobile sources are held constant from 2011 in EPA (2015). Emissions were calculated either for individual source categories or for many categories combined, using basic activity data (e.g., the amount of raw material processed or the amount of solvent purchased) as an indicator of emissions. National activity data were collected for individual categories from various agencies. Depending on the category, these basic activity data may include data on production, fuel deliveries, raw material processed, etc.

Emissions for product use were calculated by aggregating product use data based on information relating to product uses from different applications such as degreasing, graphic arts, etc. Emission factors for each consumption category were then applied to the data to estimate emissions. For example, emissions from surface coatings were mostly due to solvent evaporation as the coatings solidify. By applying the appropriate product-specific emission factors to the amount of products used for surface coatings, an estimate of NMVOC emissions was obtained. Emissions of CO and NO_x under product use result primarily from thermal and catalytic incineration of solvent-laden gas streams from painting booths, printing operations, and oven exhaust.

Activity data were used in conjunction with emission factors, which together relate the quantity of emissions to the activity. Emission factors are generally available from the EPA's Compilation of Air Pollutant Emission Factors, AP-42 (EPA 1997). The EPA currently derives the overall emission control efficiency of a source category from a variety of information sources, including published reports, the 1985 National Acid Precipitation and Assessment Program emissions inventory, and other EPA databases.

Uncertainty and Time-Series Consistency

Uncertainties in these estimates are partly due to the accuracy of the emission factors and activity data used. A quantitative uncertainty analysis was not performed.

Methodological recalculations were applied to the entire time-series to ensure time-series consistency from 1990 through 2013. Details on the emission trends through time are described in more detail in the Methodology section, above.