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**Key Points:**
- Industry-specific $C_2F_6/CF_4$ ratios are derived from atmospheric measurements
- These ratios are used to partition global total emissions to each industry
- Semiconductor and China's aluminum industry emissions most likely underestimated

**Supporting Information:**
- Readme
- Table S1

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# Quantifying aluminum and semiconductor industry perfluorocarbon emissions from atmospheric measurements

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**Abstract** The potent anthropogenic perfluorocarbon greenhouse gases tetrafluoromethane ($CF_4$) and hexafluoroethane ($C_2F_6$) are emitted to the atmosphere mainly by the aluminum and semiconductor industries. Global emissions of these perfluorocarbons (PFCs) calculated from atmospheric measurements are significantly greater than expected from reported national and industry-based emission inventories. In this study, in situ measurements of the two PFCs in the Advanced Global Atmospheric Gases Experiment network are used to show that their emission ratio varies according to the relative regional presence of these two industries, providing an industry-specific emission "signature" to apportion the observed emissions. Our results suggest that underestimated emissions from the global semiconductor industry during 1990–2010, as well as from China's aluminum industry after 2002, account for the observed differences between emissions based on atmospheric measurements and on inventories. These differences are significant despite the large uncertainties in emissions based on the methodologies used by these industries.

## 1. Introduction

The perfluorocarbons (PFCs) tetrafluoromethane ($CF_4$, PFC-14) and hexafluoroethane ($C_2F_6$, PFC-116) are among the longest-lived greenhouse gases known, with atmospheric lifetimes of about 50,000 and 10,000 years, and global warming potentials relative to carbon dioxide (100 year time scale, per unit mass of emission) of 6630 and 11,100, respectively [*Myhre et al.*, 2013]. Significant increases in atmospheric concentrations of both of these PFCs [*Mühle et al.*, 2010] are ascribed mainly to emissions from primary aluminum (AL) production during the so-called "anode events" (AEs), when the alumina feed to or within the reduction cell is restricted [*Holiday and Henry*, 1959; *International Aluminum Institute (IAI)*, 2011] and to the microchip-manufacturing component of the semiconductor (SC) industry [*Tsai et al.*, 2002; *Illuzzi and Thewissen*, 2010]. Emissions of these PFCs are included in the basket of atmospheric trace gases (carbon dioxide, methane, nitrous oxide, hydrofluorocarbons, PFCs, sulfur hexafluoride, and nitrogen trifluoride) regulated under the Kyoto Protocol of the United Nations Framework Convention on Climate Change (UNFCCC; http://unfccc.int/kyoto_protocol/items/2830.php). Both the AL and SC industries have launched voluntary efforts to control their emissions of these substances, reporting success in meeting their goals [*Illuzzi and Thewissen*, 2010; *IAI*, 2011; *World Semiconductor Council*, 2011]. Other emission sources for these PFCs are known, such as $CF_4$ released during the production of $SF_6$ and HCFC-22 [*Institute for Environmental Protection and Research*, 2013], and of $C_2F_6$ for its use as a refrigerant (the R-508 series of refrigerants contain 55–60% of $C_2F_6$). PFC emissions from these sources are estimated to be small compared to the emissions from the AL and SC industries [*Mühle et al.*, 2010; *Emissions Database for Global Atmospheric Research* (*EDGAR*), 2012]. There are very small natural emissions of $CF_4$, sufficient to maintain the preindustrial atmospheric burden [*Deeds et al.*, 2008; *Mühle et al.*, 2010]. These other sources are insignificant compared to the two main industrial sources, and they are not considered further here.

Despite the industry's efforts to reduce PFC emissions, significant fractions of the global "top-down" emissions of these two PFCs derived from atmospheric measurements remain unaccounted for in the "bottom-up" emissions reported by the industry, as has been shown by *Mühle et al*. [2010] using atmospheric measurements from the Advanced Global Atmospheric Gases Experiment (AGAGE) network [*Prinn et al*., 2000]. Mühle et al. did not, however, directly identify the sources of these top-down versus bottom-up discrepancies.

In this study, we present a method for apportioning the global total emissions of $CF_4$ and $C_2F_6$ to the AL and SC industries based on industry-representative $C_2F_6/CF_4$ emission ratios derived from AGAGE measurements in regions where one or the other industry is prevalent. These results are used to address the source of the missing emissions in the bottom-up estimates during 1990–2010 and to suggest key areas of the industry bottom-up methodology that may have caused these discrepancies.

## 2. Methods

### 2.1. Measurements of AL and SC $C_2F_6/CF_4$ Emission Ratios

The $C_2F_6/CF_4$ emission ratios for the AL and SC industries were measured at AGAGE stations, where the emissions from each industry can be clearly isolated and defined. For the AL industry, we use the AGAGE measurements at Cape Grim (Tasmania, Australia) and at Aspendale (Victoria, Australia) [*Mühle et al*., 2010; *Fraser et al*., 2011] from July 2004 to May 2010, using an air mass back trajectory analysis to identify emission plumes from two aluminum smelters (Portland and Point Henry) located in southern Australia [*Fraser et al*., 2011], where SC manufacturing does not exist. For the SC industry, we use the AGAGE measurements at Gosan Station (Jeju Island, South Korea) [*Kim et al*., 2010; *Li et al*., 2011a] from January 2008 to December 2010, using an air mass back trajectory analysis to characterize emission plumes from South Korea, Japan, and Taiwan [*Li et al*., 2011a], where only very limited AL production occurs. The SC production capacity in these three countries comprised about 58% of the global production in 2011 [*IC Insights*, 2011], and PFC emissions in these countries are almost entirely from the SC industry [*Greenhouse Gas Inventory and Research Center of Korea, Ministry of Knowledge Economy of Korea*, 2011, 2012; *Greenhouse Gas Inventory Office of Japan*, 2012; *U.S. Environmental Protection Agency (EPA)*, 2012]. Emission ratios are then calculated from enhancements above statistically determined background conditions in parts per trillion (ppt) at each measurement site [e.g., *O'Doherty et al.,* 2001]. All measurements were made in situ using custom "Medusa" gas chromatography–mass spectrometry instruments based on a cryofocusing technique [*Miller et al*., 2008] and calibrated on the AGAGE SIO-2005 scale for $CF_4$ and the AGAGE SIO-2007 scale for $C_2F_6$ [*Mühle et al*., 2010]. All ratios are derived using a bivariate fit method [*Cantrell*, 2008] to take into account measurement uncertainties in both $CF_4$ and $C_2F_6$.

These emission ratios were converted from molar concentration ratios ($0.067 \pm 0.008$ ppt/ppt for $R_{AL}$ (Figure 1a) and $0.255 \pm 0.802$ ppt/ppt for $R_{SC}$ (Figure 1b)) to mass ratios (kg/kg) by multiplying by the $C_2F_6$:$CF_4$ molecular weight ratio (1.57). The resulting mean $R_{AL}$ value is $0.10 \pm 0.01$ kg/kg based on the fit to the combined data from the two Australian smelters (converted from Figure 1a), and the mean $R_{SC}$ value is $0.40 \pm 0.19$ kg/kg (converted from Figure 1b) based on the averaged emission ratios for South Korea ($0.35 \pm 0.01$ kg/kg), Japan ($0.51 \pm 0.05$ kg/kg), and Taiwan ($0.34 \pm 0.05$ kg/kg) with each country's factor weighted by its reported SC manufacturing capacity [*IC Insights*, 2011]. The given uncertainty for $R_{AL}$ is twice the standard error in the bivariate fit, while the given uncertainty for $R_{SC}$ is twice the standard deviation of the three national emission ratios.

Our PFC emission ratio for the AL industry is in good agreement with previous estimates, including measurements of smelter-impacted air samples from the early 1970s that consistently suggest an AL industry mass emission ratio of 0.1 kg/kg [*Mühle et al*., 2010, and references therein], essentially identical to our modern-day fitted value. Our ratio also agrees, within uncertainty, with the global AL industry bottom-up reported emission ratios [*IAI*, 2011] for 2008–2010 of $0.09 \pm 0.01$ kg/kg and 1990–2010 of $0.12 \pm 0.02$ kg/kg, as well as an emission ratio of $0.11 \pm 0.02$ kg/kg measured at another Australian smelter (Kurri Kurri) by stack sampling of smelter exhaust gases [*Fraser et al*., 2013].

For the SC industry, our top-down emission ratios for South Korea of $0.35 \pm 0.01$ kg/kg and Japan of $0.51 \pm 0.05$ kg/kg are close to the bottom-up industry-reported emission ratios [*Greenhouse Gas Inventory*

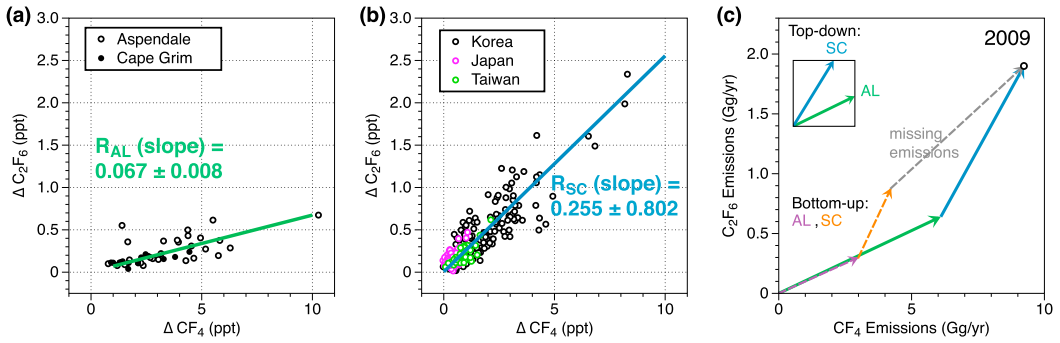


**Figure 1.** (a and b) Partitioning global PFC emissions using industry-specific $C_2F_6/CF_4$ emission ratios, $R$, derived from atmospheric measurements of emissions from regions where only one type of industry is the overwhelming PFC emitter. Aluminum industry (AL) emission ratio, $R_{AL}$, is based on in situ measurements at Cape Grim, Tasmania, and Aspendale, Victoria, Australia (Figure 1a). Semiconductor industry (SC) emission ratio, $R_{SC}$, is based on in situ measurements of South Korea, Japan, and Taiwan emissions at Gosan, Jeju Island, Korea (Figure 1b). The figure shows enhancements above local background in parts per trillion (ppt). These vector slopes are used to partition the annual global top-down $CF_4$ and $C_2F_6$ emissions between the AL and SC industries, for example, 2009. The concentration ratios (0.067 ± 0.008 ppt/ppt for $R_{AL}$ and 0.255 ± 0.802 ppt/ppt for $R_{SC}$) are converted to mass ratios (kg/kg) by multiplying 1.57 and shown as vector slopes in the inset of Figure 1c. The green and blue vectors represent the global top-down PFC emissions from the AL and SC industries for that year, with the end point at the global total top-down emissions. The bottom-up global PFC emission vectors for 2009 are also shown in Figure 1c in purple and orange for the AL and SC industries, respectively, and the global top-down versus bottom-up discrepancies are shown in gray.

*and Research Center of Korea*, 2011–2012; *Greenhouse Gas Inventory Office of Japan*, 2012] for 2008–2010 of 0.34 ± 0.04 kg/kg and 0.45 ± 0.07 kg/kg, respectively. This gives some credence to the agreement, within combined uncertainties, between our top-down $R_{SC}$ of 0.40 ± 0.19 kg/kg and the global SC industry bottom-up emission ratios derived in this study (section 2.3 and in the Appendix A) of 0.51 ± 0.01 kg/kg for 2008–2010 and 0.64 ± 0.12 kg/kg for 1990–2010. Nonetheless, significant uncertainties remain in our $R_{SC}$, in part due to lack of knowledge of SC industry emission ratios in regions outside East Asia.

To account for the increased uncertainties in our fitted $R_{AL}$ and $R_{SC}$ prior to 2008, where we lack data for a more thorough uncertainty analysis, we apply a percentage uncertainty for 1990 that is double our uncertainty for the period 2008–2010 and linearly interpolate these uncertainties for the period 1990–2008.

### 2.2. Global Proportioning of the PFC Emissions to AL and SC Industries

We define the annual top-down global PFC emissions $E_{CF4}$ and $E_{C2F6}$ as the sum of annual global $CF_4$ and $C_2F_6$ emissions from each industry:

$$E_{CF4} = E_{CF4:AL} + E_{CF4:SC} \quad (1)$$

$$E_{C2F6} = E_{C2F6:AL} + E_{C2F6:SC} \quad (2)$$

where the SC and AL additions to the subscripts refer to the two respective industries. By applying our mean $R_{AL}$ and $R_{SC}$ industry-specific emission ratios to equation (2), we obtain

$$E_{C2F6} = E_{CF4:AL} \times R_{AL} + E_{CF4:SC} \times R_{SC} \quad (3)$$

Equations (1) and (3) are then solved for the variables $E_{CF4:AL}$ and $E_{CF4:SC}$, from which the industry-specific $C_2F_6$ emissions are calculated. This method, shown in vector form in Figure 1c for 2009, is applied to each year in the period 1990–2010 to obtain the time series of industry-specific top-down global emissions for $CF_4$ and $C_2F_6$.

While we account for various sources of uncertainties in our method, including uncertainties in the $R_{AL}$ and $R_{SC}$ explained above, we find that overall, the uncertainties in our top-down industry-specific emissions are dominated by the uncertainties of the global top-down emission estimates.

### 2.3. Updating the Global Top-Down and Bottom-Up Emissions of $CF_4$ and $C_2F_6$

To improve our estimates of the global top-down versus bottom-up discrepancies in $CF_4$ and $C_2F_6$ emissions, we compiled the latest industry and government reporting of their bottom-up emissions. For the AL industry,

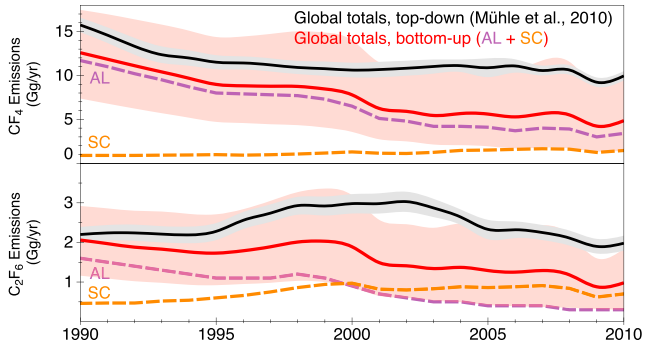


**Figure 2.** Global discrepancies between top-down and bottom-up PFC emission estimates. The bottom-up reported emissions of $CF_4$ and $C_2F_6$, shown in red, are the sum from the two main sources, the aluminum (AL) and semiconductor (SC) industries, shown in purple and orange, respectively, and account for only two thirds to one half of the measurement-derived top-down emissions of these compounds, which are shown in black with their uncertainties in gray. AL industry bottom-up emissions are taken from *IAI* [2011]. SC industry bottom-up emissions have been compiled in this study from various reported sources, as described in Appendix A. Uncertainties of the global bottom-up emission, shown in light red, are the sum of the uncertainties from the AL and SC industries, displayed individually in Figure 3. The discrepancies between the top-down and bottom-up emissions increase in time, especially since 2001, revealing increasing uncertainties in the bottom-up emission inventories.

the International Aluminum Institute (IAI) [*IAI*, 2011] has recently updated their emission estimates for the global AL industry based on new results from China, which we incorporate into our study. Uncertainties in the AL industry inventories are based on Monte Carlo analysis performed by the IAI for 1990 and 2010 [*IAI*, 2011], which we interpolate for the years in between.

PFC emissions from the SC industry reported by the World Semiconductor Council [*World Semiconductor Council*, 2011] are not as specifically defined as they are reported as a single sum of all fluorinated gases emitted during manufacture rather than as compound-specific emissions. For this study, we derive a new bottom-up SC estimate based on the National Inventory Reports (NIRs) to the United Nations Framework Convention on Climate Change (UNFCCC; http://unfccc.int/national_reports/annex_i_ghg_inventories/national_inventories_submissions/items/6598.php) and on reported global anthropogenic non-$CO_2$ greenhouse gas emissions during the period 1990–2030 [*EPA*, 2012]. The Emissions Database for Global Atmospheric Research (EDGAR)[*EDGAR*, 2012], an emission inventory widely cited in many studies, was not used for this study due to various concerns regarding its reporting, including unrealistically low PFC emissions estimated for South Korea. Details of our bottom-up methodology are given in Appendix A.

## 3. Results and Discussions

### 3.1. Global Discrepancies in PFC Emissions, Top-Down Versus Bottom-Up

Comparison of the top-down emissions, updated from *Mühle et al.* [2010] to 2010, against the updated bottom-up emissions of $CF_4$ and $C_2F_6$ are shown in Figure 2. Our results show that for the period 1990–2010, about one third (34% of $CF_4$, 35% of $C_2F_6$, and the black curve compared to the red curve) of the global top-down emissions are unaccounted for in the bottom-up emission inventories. Furthermore, the discrepancy increases to almost one half (50% for $CF_4$ and 48% for $C_2F_6$) for the 2002–2010 part of this period, suggesting that uncertainties in the bottom-up emissions have increased in recent years. In the following sections, we compare the top-down and bottom-up emissions from each industry to better understand the source(s) of these discrepancies.

### 3.2. SC Industry Emissions

Our results for industry-specific top-down PFC emissions, shown against their respective bottom-up counterparts in Figure 3, suggest that the SC industry is the dominant source of discrepancy in the global PFC budgets. Averaged over 1990–2010, we find that the reported SC industry bottom-up emissions are ~3.6 (1.4 to 25) times lower than the top-down values for $CF_4$ and ~2.4 (0.88 to 8.4) times lower for $C_2F_6$. The discrepancy in the SC industry emissions is shown to have peaked in 2002, suggesting that improvements in manufacturing technologies may have had a significant impact on reducing PFC emissions in later years,

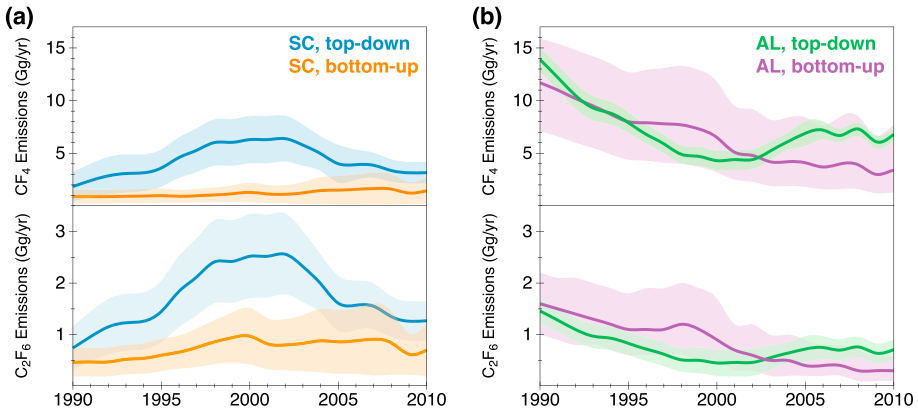


**Figure 3.** Comparisons of top-down global $CF_4$ and $C_2F_6$ emissions for (a) the semiconductor industry, SC, and for (b) the aluminum industry, AL, obtained by the methods described in equations (1)–(3) and illustrated in Figure 1c, against the bottom-up emissions from the SC and AL industries described in section 2.3 and Appendix A. The shaded areas show the uncertainties used in the calculations (described in section 2.2) in their respective colors. The largest discrepancies between bottom-up and top-down emissions are found for the semiconductor industry in the middle years of this study and for the aluminum industry after 2002.

notably a gradual replacement of $C_2F_6$ with nitrogen trifluoride [*Fthenakis et al.*, 2010; *Illuzzi and Thewissen*, 2010; *Arnold et al.*, 2013]. However, significant sources of uncertainty remain unaddressed in the reported bottom-up emission inventories, particularly whether the UNFCCC emission factors used to calculate emissions [*Bartos et al.*, 2006] actually apply to the wide range of real-life manufacturing conditions in the SC industry [*Kastenmeier*, 1996; *International Semiconductor Manufacturing Technology Manufacturing Initiative*, 1997; *Koike et al.*, 1997; *Namose*, 2003; *Kuroki et al.*, 2005]. Another uncertainty is the wide range of actual efficiencies in PFC removal ("abatement") techniques that are currently widely applied in SC factory exhaust systems, where the assumed 90% abatement efficiency [*Bartos et al.*, 2006] is likely to overstate real-world abatement rates [*Czerniak et al.*, 2007; *Ou Yang et al.*, 2009; *Choi et al.*, 2012]. Details of SC industry bottom-up emission accounting are generally protected as trade secrets, which make a more thorough analysis of these uncertainties problematic.

### 3.3. AL Industry Emissions

For the AL industry, we find general agreement within uncertainties between top-down and bottom-up emissions, giving independent support to the substantial emission reductions reported by the industry [*IAI*, 2011]. However, after 2001, the gradually increasing trend we find in AL industry top-down emissions is inconsistent with the gradually decreasing industry-reported values, which correspond with the increase in top-down versus bottom-up discrepancies observed in the global emissions (Figure 2). During this period, aluminum production in China also increased markedly, from 11% of global production in 2000 to 39% in 2010 [*IAI*, 2011]. The AL industry's PFC emission estimates for China are reported to be more uncertain, because detailed records of manufacturing conditions used in the more accurate emission accounting typically used by the IAI outside of China are not available for most/many Chinese smelters. Instead, a single-emission factor derived from a survey of a small number of facilities in China is used for all Chinese production [*IAI*, 2011; *Li et al.*, 2011c, 2012]. Note that this factor (0.69 t PFC $CO_2$ e/t Al) was found to be significantly larger than the global industry average outside of China (0.23 t PFC $CO_2$ e/t Al). The AL industry's own PFC emission estimates for China (1.37 Gg/yr $CF_4$, 0.06 Gg/yr $C_2F_6$, 2008–2010 average), based on this larger emission factor, are still lower than the top-down estimates of 2.2 (1.4 to 3.1) Gg/yr $CF_4$ for 2007–2008, and 0.5 (0.4 to 0.7) Gg/yr $C_2F_6$ for 2007–2009 found in previous studies [*Kim et al.*, 2010; *Saito et al.*, 2010; *Li et al.*, 2011a].

Given China's relatively small footprint in the global SC industry of 9% in 2011 [*IC Insights*, 2011], the mismatch between China's top-down and bottom-up numbers most likely suggests that AL bottom-up emissions for China remain significantly underestimated. The AL industry is currently working to better define its bottom-up estimates of Chinese PFC emissions [*Li et al.*, 2011b, 2011c, 2012; *Marks and Bayliss*,

2012; *Chen et al*., 2013] and is also implementing various emission-reducing technologies to their manufacturing processes [*Li et al*., 2013]. In addition, work is ongoing to better define PFC emissions during cell start-up procedures, anode changes, and other non-AE periods. These effects are typically not included in the current AL industry bottom-up estimates, as they have just recently been described in the literature [*Maltais et al*., 2010; *Li et al*., 2011b; *Marks and Bayliss*, 2012; *Chen et al*., 2013], and may be particularly significant for the very large reduction cells predominantly used in China. Their inclusion should lead to better defined and subsequently reduced PFC emissions from the Chinese AL industry.

## Appendix A: Bottom-Up Emissions From the SC Industry

The use of the National Inventory Reports (NIRs) for deriving a bottom-up emission inventory of the SC industry is not straightforward, as each country's NIR report their emissions in different formats, and NIRs for some countries are not available. Here we describe our methods for aggregating the available information to derive a SC bottom-up inventory for this study.

For countries that report compound-specific emissions in their NIRs to UNFCCC, namely, the U.S., Belgium, Germany, France, Ireland, Italy, Sweden, Czech Republic, Switzerland, Canada, and Russia, we directly use the $CF_4$ and $C_2F_6$ emissions reported in the NIRs directly.

Some European countries, namely, Netherlands and Malta, choose to report their SC emissions aggregated for all the PFC species (including $CF_4$, $C_2F_6$, $C_3F_8$, and c-$C_4F_8$). To derive compound-specific emissions from this total, we use the per-compound emission distribution found for other European countries to estimate the fraction of emissions of $CF_4$ and $C_2F_6$, assuming that emission patterns among European countries are similar. Emissions for the U.S. and EU are reported on a yearly basis for 1990–2010, which we used directly in our study.

Emissions for Japan [*Greenhouse Gas Inventory Office of Japan*, 2012] and South Korea [*Greenhouse Gas Inventory and Research Center of Korea, Ministry of Knowledge Economy of Korea*, 2011, 2012] are reported aggregated for all the PFCs for 1995–2010; however, both countries make available consumed amounts of individual compounds for some years (1995, 2000, and 2002–2010 for Japan and 2007–2010 for South Korea), allowing us to make compound-specific emissions in these years based on the most recent 2006 Intergovernmental Panel on Climate Change (IPCC) bottom-up methodology [*Bartos et al*., 2006]. This is a deviation from Japan's NIR, which uses older emission factors from 2002 [*Bartos and Burton*, 2002] for their calculations, and our update makes Japan's emissions more comparable to those from other countries that have adopted the IPCC 2006 emission factors. For years between 1995 and 2006, where consumption data are not available, we either perform linear interpolation from bracketing years (Japan) or proportional to the reported aggregated PFC emissions (South Korea). Emissions prior to 1995 for both countries are assumed to be at 1995 levels.

One additional correction necessary for estimating Japan's emissions is estimating the percentage of abatement (removal of PFCs in the factory exhaust) applied at the semiconductor factories, which is not directly specified in Japan's NIR. We do this by comparing the total aggregated PFC emissions of our compound-specific emission inventory (presumably, the totals before abatement) against the reported total aggregated emissions in Japan's NIRs (the totals after abatement). From these comparisons, we estimate no abatement prior to 2003 and exponentially increasing abatement since then to 47% in 2010. Such corrections are not necessary for South Korea, which does not abate its emissions.

For countries where NIRs are not available, namely, China, Taiwan, and countries in Southeast Asia such as Singapore, we scaled our South Korean emissions based on the aggregated PFC emissions reported for these countries in the U.S. Environmental Protection Agency (EPA) on global anthropogenic non-$CO_2$ greenhouse gas emissions during 1990–2030 [*EPA*, 2012]. The emissions for these countries and South Korea in the EPA report are all based on country-specific SC manufacturing capacity shares, which make the EPA emissions a good proxy for the relative emissions among these countries. The estimates in the EPA report are only available for every 5 year increment from 1990 to 2010, and therefore, yearly emissions were interpolated between the reported years.

Uncertainties derived for the SC bottom-up estimates are based directly on those reported in the NIRs, for example, 20% for the U.S., 0.31% for EU, and 64% for Japan. Japan's uncertainty is most detailed, applying 50%

uncertainty in the emission factors and 40% uncertainty in the activity data to derive its total uncertainty of 64%. The Korean NIR also lists 50% uncertainty for emission factors but does not explicitly explain its uncertainty in activity data; as such, we adopt Japan's uncertainty of 64% for South Korea as well. For uncertainties in regions with no NIRs, we double the uncertainty of the highest reported uncertainties (Japan's 64%), which we assume accounts for the uncertainties due to the lack of specific data from these regions, for example the uncertainty in assuming that the emission characteristics are comparable to those of South Korea.

**Acknowledgments**

AGAGE measurement data, shown in Figures 1a and 1b, are available at the Carbon Dioxide Information Analysis Center (http://cdiac.esd.ornl.gov/ndps/alegage.html). Data supporting Figures 1c, 2, and 3 are available in Table S1 in the supporting information. The AGAGE research program is supported by the NASA Upper Atmospheric Research Program in the U.S. with grants NNX11AF17G to MIT, NNX11AF15G and NNX11AF16G to SIO, and by CSIRO and the Australian Government Bureau of Meteorology in Australia. Measurements at Gosan were supported by the Korea Meteorological Administration Research and Development Program under grant CATER 2012–3010. We would like to thank Jerry Marks for his constructive discussions regarding the aluminum industry, especially in China. We also acknowledge the strong support provided by field operators Kyeong-Sik Kang, Minkook Han (for Gosan), Jeremy Ward, Nigel Somerville (Cape Grim), and the late Laurie Porter (Cape Grim and Aspendale).

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