



## Regional atmospheric emissions determined from measurements at Jeju Island, Korea: Halogenated compounds from China

Jooil Kim,<sup>1</sup> Shanlan Li,<sup>1</sup> Kyung-Ryul Kim,<sup>1,2</sup> Andreas Stohl,<sup>3</sup> Jens Mühle,<sup>4</sup> Seung-Kyu Kim,<sup>1</sup> Mi-Kyung Park,<sup>1,2</sup> Dong-Jin Kang,<sup>5</sup> Gangwoong Lee,<sup>6</sup> Christina M. Harth,<sup>4</sup> Peter K. Salameh,<sup>4</sup> and Ray F. Weiss<sup>4</sup>

Received 16 March 2010; revised 19 April 2010; accepted 23 April 2010; published 16 June 2010.

[1] High-frequency in-situ measurements of a wide range of halogenated compounds including chlorofluorocarbons (CFCs), halons, hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs), perfluorinated compounds (PFCs), sulfur hexafluoride (SF<sub>6</sub>), and other chlorinated and brominated compounds have been made at Gosan (Jeju Island, Korea). Regional emissions of HCFC-22 (CHClF<sub>2</sub>) calculated from inverse modeling were combined with interspecies correlation methods to estimate national emissions for China, a major emitter of industrial halogenated gases. Our results confirm the signs of successful phase-out of primary ozone-depleting species such as CFCs, halons and many chlorinated or brominated compounds, along with substantial emissions of replacement HCFCs. Emissions derived for HFCs, PFCs, and SF<sub>6</sub> were compared to published estimates and found to be a significant fraction of global totals. Overall, Chinese emissions of the halogenated compounds discussed here represent 19(14–17)% and 20(15–26)% of global emissions when evaluated in terms of their Ozone Depletion Potentials and 100-year Global Warming Potentials, respectively. **Citation:** Kim, J., et al. (2010), Regional atmospheric emissions determined from measurements at Jeju Island, Korea: Halogenated compounds from China, *Geophys. Res. Lett.*, 37, L12801, doi:10.1029/2010GL043263.

### 1. Introduction

[2] Halogenated organic compounds are used in many commercial and industrial applications including refrigeration, air conditioning, foam blowing, and fire extinguishing and also as chemical solvents. They are of great concern due to their stratospheric ozone depleting potential (ODP) [Clerbaux and Cunnold, 2007]. The Montreal Protocol has been instrumental in imposing a phase-out of primary ozone-depleting substances (ODSs) such as CFCs, halons, and other long-lived chlorinated or brominated compounds (e.g.,

CH<sub>3</sub>CCl<sub>3</sub>, CH<sub>3</sub>Br). Most developed countries have already moved the majority of their consumption to HFCs, which do not destroy stratospheric ozone, driving the atmospheric decline for the most potent ODSs [Clerbaux and Cunnold, 2007; Prinn et al., 2000]. However, developing countries have been placed under a less demanding phase-out schedule, with consumption allowed for interim replacement HCFCs with reduced ODP until 2030 [United Nations Environment Programme, 2009]. As a consequence the recent rise in atmospheric HCFCs is likely caused by the emissions from developing countries [Montzka et al., 2009]. HFCs, while posing no threat to stratospheric ozone, are nevertheless of concern because of their high GWP [Forster et al., 2007], with emissions of HFCs projected to equal 9–19% (in CO<sub>2</sub>-equation basis) of CO<sub>2</sub> emissions by the year 2050 [Velders et al., 2009].

[3] Emissions of anthropogenic halogenated compounds from China are of special interest, due to the large emissions expected from its fast growing economy. In compliance with the Montreal Protocol, China has implemented a strategy to accelerate the phase-out of primary ODSs (UNEP Multilateral Fund, Phase-out plans and projects, www.multilateralfund.org/files/Policy55Plans.pdf, as of April 2008). Recent reports [Vollmer et al., 2009; Wan et al., 2009] show that China's efforts to decrease emissions of primary ODSs have been successful, leading to increased emissions of replacement HCFCs. However, some disagreements remain concerning the actual emission rates. For some HFCs, Chinese emissions have been estimated for a few species based on limited sampling of elevated-concentration “pollution events” of Chinese origin at Hateruma Island east of Taiwan [Yokouchi et al., 2006], however emissions for many HFCs remain poorly quantified. Chinese emissions of PFCs and SF<sub>6</sub> are expected to be large fractions of the global emissions [Emission Database for Global Atmospheric Research (EDGAR), 2009; Vollmer et al., 2009].

[4] In this study, we estimate Chinese emissions of 21 anthropogenic halogenated compounds for 2008 based on “top-down” interpretation of atmospheric high-precision in-situ measurements at Gosan station (126.17°E, 33.28°N), a remote island site on Jeju Island in the Yellow Sea south of the Korean peninsula, which has been shown to be an important site for understanding atmospheric emissions from East Asia [Bush and Valero, 2003; Nakajima et al., 2007]. We report emission estimates for CFC-11 (CCl<sub>3</sub>F), CFC-12 (CCl<sub>2</sub>F<sub>2</sub>), CFC-114 (CClF<sub>2</sub>CClF<sub>2</sub>), HCFC-22 (CHClF<sub>2</sub>), HCFC-141b (CH<sub>3</sub>CCl<sub>2</sub>F), HCFC-142b (CH<sub>3</sub>CClF<sub>2</sub>), HFC-23 (CHF<sub>3</sub>), HFC-134a (CH<sub>2</sub>FCF<sub>3</sub>), HFC-152a (CH<sub>3</sub>CHF<sub>2</sub>),

<sup>1</sup>School of Earth and Environmental Sciences, Seoul National University, Seoul, South Korea.

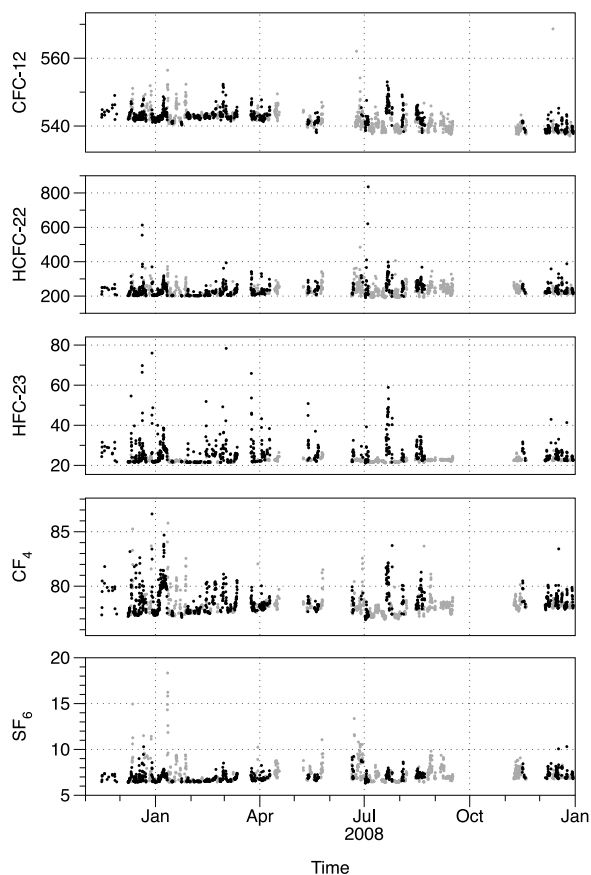
<sup>2</sup>Research Institute of Oceanography, Seoul National University, Seoul, South Korea.

<sup>3</sup>Norwegian Institute for Air Research, Kjeller, Norway.

<sup>4</sup>Scripps Institution of Oceanography, University of California, San Diego, La Jolla, California, USA.

<sup>5</sup>Korea Ocean Research and Development Institute, Seoul, South Korea.

<sup>6</sup>Environmental Science Division, Hankuk University of Foreign Studies, Yongin, South Korea.



**Figure 1.** Time series of measurements at Gosan station for CFC-12, HCFC-22, HFC-23, CF<sub>4</sub>, and SF<sub>6</sub>. Data collected during atmospheric transport from China are shown in black, while other data are shown in gray. We mostly detect Chinese pollution signals in the winter season, when the monsoon winds favor transport of air masses from the north-northwest to Gosan station.

HFC-32 (CH<sub>2</sub>F<sub>2</sub>), HFC-125 (CHF<sub>2</sub>CF<sub>3</sub>), HFC-143a (CH<sub>3</sub>CF<sub>3</sub>), H-1211 (CBrClF<sub>2</sub>), CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub>, SF<sub>6</sub>, CH<sub>3</sub>Cl, CH<sub>2</sub>Cl<sub>2</sub>, CHCl<sub>3</sub>, and CH<sub>3</sub>Br.

## 2. Methods

### 2.1. Measurement and Data Selection

[5] At Gosan ambient concentrations of halogenated compounds (time series of selected species are shown in Figure 1) are measured in 2 L air samples with the “Medusa” automated custom-built gas chromatographic system with mass spectrometric detection [Miller *et al.*, 2008], with relative measurement precisions typically less than 1%. Our measurements are closely tied to the Advanced Global Atmospheric Gases Experiment (AGAGE) [Prinn *et al.*, 2000], and are reported as dry air mole fractions on the calibration scales developed at the Scripps Institution of Oceanography (SIO), and the University of Bristol (UB) [Prinn *et al.*, 2000]: SIO-98 (CHCl<sub>3</sub>), SIO-07 (HFC-23, -32, -143a, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub>), UB-98 (HFC-125, CH<sub>2</sub>Cl<sub>2</sub>), SIO-05 (all other compounds). The measurements used here were made from November 2007 to December 2008 at Gosan station, Jeju Island, Korea. Gaps in the data arise from difficulties in maintaining continuous operation of the instrumentation, and

are a source of increased uncertainty in our emission calculations (detailed in section 2.2).

[6] A Lagrangian particle dispersion model (FLEXPART) [Stohl *et al.*, 2005] was used to identify periods of air transport from China, resulting in 825 data points (29% of all measurements) to derive emissions from China. Large-scale meteorology at Gosan is dominated by the East Asian Monsoon and the majority of air transport events from China occur in winter (59% of all Chinese data points) with wind from north-northwest. Our emission results are not expected to be affected by this seasonal sampling distribution for most species since emissions from the industrial and commercial sources for these species are expected to be fairly constant throughout the year. In support of this assumption, we find that interspecies ratios do not vary seasonally, as shown in the auxiliary material.<sup>1</sup>

### 2.2. Emissions Estimation

[7] Emissions of the measured halogenated compounds were derived using an interspecies correlation technique, whereby the correlation of a target compound *x* with a tracer compound *y* is converted into an emission rate of the target compound *x* by scaling the target/tracer ratio *x/y* to the emissions of the tracer *y* [e.g., Yokouchi *et al.*, 2006]. For this study, HCFC-22 was chosen as the main tracer *y* because it showed statistically significant ( $p < 0.01$ ) correlation with most other species. Pearson correlation coefficients between all species are available in the Figure S3 of the auxiliary material. In China HCFC-22 is used mainly for air-conditioning and refrigeration [Wan *et al.*, 2009], and the widespread distribution of its sources may partly explain these good correlations.

[8] It is important to note that this interspecies correlation approach inherently requires that the sources of the species that are compared be effectively coincident, both spatially and temporally. To the extent that this is not the case, the results obtained by this method may be biased.

[9] Emissions of HCFC-22 were calculated directly from our measurements using atmospheric inversion techniques based on FLEXPART [Stohl *et al.*, 2009], yielding estimated emissions ranging from 64 to 109 kt/a, with a best guess of 83 kt/a. Details on a priori parameters are given in the supplementary online material.

[10] Despite their relatively low, but still significant, correlations with HCFC-22 ( $r^2 = 0.21\text{--}0.29$ ), our estimates of CFC-12, CFC-114, HFC-143a, and C<sub>3</sub>F<sub>8</sub> emissions are included to show the effectiveness of Chinese phase-out of CFC-12 and CFC-114 [Wan *et al.*, 2009] and to provide a baseline for future emissions of C<sub>3</sub>F<sub>8</sub> and HFC-143a. While measurements of CFC-113 (C<sub>2</sub>F<sub>3</sub>Cl<sub>3</sub>), H-1301 (CF<sub>3</sub>Br), CH<sub>3</sub>CCl<sub>3</sub> and HFC-365mfc (CH<sub>3</sub>CF<sub>2</sub>CH<sub>2</sub>CF<sub>3</sub>) are available, emissions were not calculated for these species due to their very poor correlations against other species including HCFC-22 ( $r^2 < 0.11$ ). Emissions for CFC-113, H-1301, and CH<sub>3</sub>CCl<sub>3</sub> are currently expected to be near zero in China [Vollmer *et al.*, 2009; Wan *et al.*, 2009] while HFC-365mfc is known to be used mainly in Europe [Vollmer *et al.*, 2006], which may explain the lack of correlation.

[11] Correlations between species are analyzed based on enhancements above the ambient baseline concentration

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2010GL043263.

**Table 1.** Emissions of Halogenated Compounds From China Estimated in This Study and Previously Reported Values for Chinese and Global Emissions

Compound	China Emissions (kt/a)					Global Emissions <sup>a</sup> (kt/a)
	This Study for 2008	(% to Global)	<i>Yokouchi et al.</i> [2006] for 2005	<i>Vollmer et al.</i> [2009] for 2007	<i>Wan et al.</i> [2009] for 2008	
CFC-11	12 (9.4 – 17)	15		33 (26 – 43)	14.259	82 (±30)
CFC-12	6.3 (4.7 – 8.5)	8		14 (9 – 19)	3.869	79 (±40)
CFC-114	1.3 (0.9 – 1.8)					
HCFC-22 <sup>b</sup>	83 (64 – 109)	23	54 (±34)	165 (140 – 213)	79.268	365 (±70)
HCFC-141b	15 (12 – 21)	25			12.148	60 (±10)
HCFC-142b	10 (7.6 – 13)	24		12 (10 – 18)		41 (±5.0)
HFC-23	12 (8.6 – 15)	89	10 (±4.6)		10.6	13.5
HFC-134a	8.7 (6.5 – 12)	5	3.9 (±2.4)		1.03	165
HFC-152a	5.7 (4.3 – 7.6)	20	4.3 (±2.3)		0	28.5
HFC-32	4.3 (3.2 – 5.9)	119			0	3.6
HFC-125	3.2 (2.4 – 4.4)	16			0	20.5 (±4.3)
HFC-143a	0.59 (0.44 – 0.80)	2			0	31.7
H-1211	1.4 (1.0 – 1.9)	23		2.1 (1.7 – 2.5)	1.353	6.2 (±3.0)
CF <sub>4</sub>	2.3 (1.7 – 3.1)	26			1.46	8.92
C <sub>2</sub> F <sub>6</sub>	0.49 (0.37 – 0.66)	27			0.24	1.83
C <sub>3</sub> F <sub>8</sub>	0.09 (0.07 – 0.12)	21			0.0007	0.42
SF <sub>6</sub>	1.3 (0.93 – 1.7)	19		0.80 (0.53 – 1.10)	1.64	6.8 (±0.41)
CH <sub>3</sub> Cl	265 (200 – 354)					
CH <sub>2</sub> Cl <sub>2</sub>	176 (133 – 234)					
CHCl <sub>3</sub>	49 (38 – 66)			86 (51 – 140)		
CH <sub>3</sub> Br	5.8 (4.3 – 7.9)			0.24 (0.17 – 0.47)		

<sup>a</sup>Emissions for 2007 from *Vollmer et al.* [2009] (CFC-11, CFC-12, HCFC-22, HCFC-141b, HCFC-142b, H-1211), *Montzka et al.* [2010] (HFC-23), *O'Doherty et al.* [2009] (HFC-125) and *Levin et al.* [2010] (SF<sub>6</sub>). All others from *EDGAR* [2009] extrapolated to 2008.

<sup>b</sup>HCFC-22 emissions in our study were calculated from inverse modeling.

determined statistically using the AGAGE algorithm [*O'Doherty et al.*, 2001], thereby removing the effects of changes in the baseline itself. The enhancements over the baseline will be denoted as “delta” ( $\Delta$ ) values in this study. The error range in our emission estimates account both for uncertainties in the emissions estimates of HCFC-22 derived from our inversions and the 95% error range in the correlation slope against  $\Delta$ HCFC-22.

### 3. Results and Discussions

#### 3.1. Global Perspective

[12] To compare our 2008 Chinese emissions (Table 1 and Figure 2) with global emissions, we calculated the fraction of Chinese emissions to global emissions reported by *Vollmer et al.* [2009], *Montzka et al.* [2010], *O'Doherty et al.* [2009] (for 2007), *Levin et al.* [2010], and *EDGAR* [2009] (*EDGAR* [2009] values in this study have been extrapolated to 2008 using reported emissions from 2000–2005). Our 2008 results show that emissions of many individual species are close to or exceed 20% of global emissions. The combined emissions derived in this study are equivalent in terms of ODP [*Clerbaux and Cunnold*, 2007] to the emission of 50 kt of CFC-11 per year, or in terms of GWP (100-year time horizon [*Forster et al.*, 2007]) to the emissions of 602 Mt of CO<sub>2</sub> per year. These values represent approximately 19(14–27)% of global emissions of major halocarbon species (assuming CCl<sub>4</sub> emissions of 15 kt/a for China and 53 kt/a globally [*Vollmer et al.*, 2009]) in terms of their ODPs, and about 20(15–26)% of global emissions of major halocarbon species in terms of their GWPs (error range based on uncertainties in our results and of CCl<sub>4</sub> as reported by *Vollmer et al.* [2009], assuming that global totals are correct). These results show the importance of Chinese contributions to global

emissions of these anthropogenic halogenated species, the magnitudes of which are now close to China's 20% of global population.

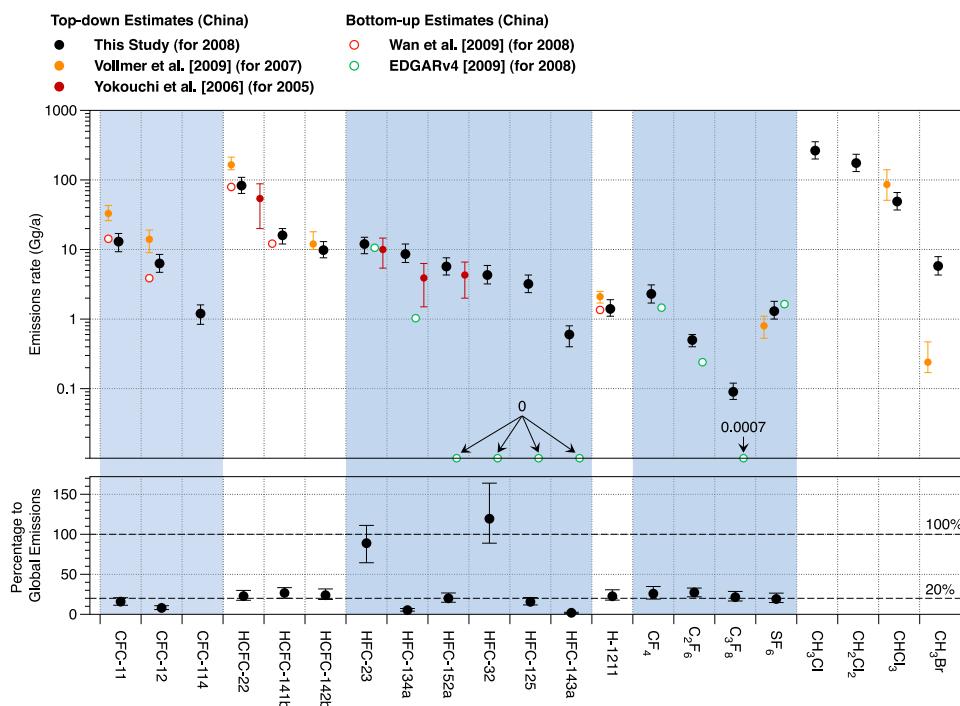
#### 3.2. Emissions of Primary ODSs and HCFCs

[13] Our 2008 emission estimates for primary ODSs and HCFCs point to decreasing emissions of primary ODSs in conjunction with dramatically rising Chinese emissions of HCFCs, in line with the findings of *Vollmer et al.* [2009] and *Wan et al.* [2009]. This confirms the overall success of China's phase-out plan for primary ODSs, and the dominant role of HCFCs in China's current consumption of halocarbons.

[14] Note that our 2008 emission estimates agree within uncertainties with the 2008 emissions projected by *Wan et al.* [2009], while being much lower than the 2007 emissions reported by *Vollmer et al.* [2009]. Discrepancies in the emissions of CFCs between our study and *Vollmer et al.* [2009] may be related to the dramatic decrease in consumption reported for CFCs in China (from 5,832.1 CFC-11 equivalent ODP tonnes in 2007 to 263.0 tonnes in 2008; data from <http://ozone.unep.org>, updated to Dec. 2009). However the nearly two-fold difference in the emissions of HCFC-22 suggests that the values of *Vollmer et al.* [2009] may be overestimated, since a decrease in this widely-used species is unlikely, especially to such degree. Differences in sampling location may explain these discrepancies, and an inversion study is in progress to address the errors of single-station emission analyses by assimilating data from several stations [*Stohl et al.*, 2010].

#### 3.3. Emissions of HFCs

[15] Our 2008 estimates of HFC-23 emissions are in good agreement with an extrapolation of Chinese emissions estimated by *EDGAR* [2009]. China's emissions of HFC-23 are



**Figure 2.** Chinese emissions derived in this study and other studies are shown in the top plot. The results from *Vollmer et al.* [2009] are mostly higher than those in our study and by *Wan et al.* [2009]. The differences between *Yokouchi et al.* [2006] and our study reflect the increase of emissions since 2005. Emissions estimated by *EDGAR* [2009] are mostly lower, especially for HFC-152a, HFC-32, HFC-125, and  $C_3F_8$ . Comparing our results to global emissions (as shown in Table 1), Chinese emissions of many species are near 20% of global emissions as shown in the lower plot.

shown to be an overwhelmingly large fraction of the global total. These emissions are expected from the production of HCFC-22, where HFC-23 is emitted as a by-product [*McCulloch and Lindley*, 2007]. Some of the HFC-23 emissions are now abated through the United Nations Clean Development Mechanism (CDM), however the continued increase in atmospheric levels of HFC-23 suggests that substantial emissions remain [*Montzka et al.*, 2010]. Ten factories in China are currently known to participate in CDM. While the relatively small increase in emissions since 2005 [*Yokouchi et al.*, 2006] might be attributable to these CDM activities, direct validation of the effects of the CDM are difficult considering the relatively large range of uncertainties in our estimates.

[16] Chinese emissions of HFC-134a have increased sharply since 2005 [*Yokouchi et al.*, 2006] and are larger than those reported by *EDGAR* [2009]. This suggests that the emissions of *EDGAR* [2009] are underestimated and/or the emission increase since 2005 was much larger than the extrapolated 2000 to 2005 trend. HFC-134a is the only HFC for which actual consumption in China is known (as a refrigerant in the mobile air conditioning sector), and should continue to increase substantially with the growth of the Chinese automotive industry [*Hu et al.*, 2009].

[17] Chinese emissions of HFC-152a, HFC-125, and especially HFC-32 contribute significantly to global emissions, while the emissions of HFC-143a are relatively small. These four compounds are mainly used in developed countries as foam blowing agents, aerosol propellents, and in refrigeration mixtures [*Velders et al.* 2009]. Chinese emissions of these compounds are reported to be zero throughout 2000–2005 by *EDGAR* [2009], in line with the understanding

that actual Chinese consumption for these species is currently minimal (D. Wan, personal communication, 2009). While the emissions of HFC-152a, HFC-32, and HFC-125 may have increased substantially since 2005, the 2005 HFC-152a emissions reported by *Yokouchi et al.* [2006] suggest that the emissions of *EDGAR* [2009] are significantly underestimated, at least for HFC-152a. Global emission estimates for these compounds may also need to be revised to better account for emissions from China. Fugitive leaks during production may be one possible explanation, but more research is needed to clearly define the emission sources for these compounds, especially given the global importance of their emissions. Emission patterns inferred from interspecies ratios of HFC-152a, HFC-32, and HFC-125 are discussed further in the supplementary online material.

### 3.4. Emissions of PFCs and $SF_6$

[18] Emissions of  $CF_4$  and  $C_2F_6$  derived in our study for 2008 are consistent with emissions of *EDGAR* [2009], but our emissions for  $C_3F_8$  are much larger. These PFCs are emitted mostly from aluminum smelting ( $CF_4$ ,  $C_2F_6$ ), semiconductor industries ( $CF_4$ ,  $C_2F_6$ ,  $C_3F_8$ ), and refrigeration/air conditioning ( $C_3F_8$ ), and are of interest due to their long atmospheric lifetimes (several 1000 to 10000s of years) that lead to an almost permanent burden to the radiative budget of the atmosphere [*Forster et al.*, 2007]. The large fractions of Chinese emissions of global totals of these compounds reflect the active aluminum smelting and electronics industries in China. *Mühle et al.* [2010] have recently refined the atmospheric histories and global emissions of these three PFCs.

[19] Our 2008 emission estimate for  $SF_6$  agrees within uncertainties with the emissions reported by *Vollmer et al.*

[2009] and EDGAR [2009] and confirms the substantial contribution of China's SF<sub>6</sub> emissions to global totals. Mainly used as insulating material in high-voltage electrical equipment, SF<sub>6</sub> is a powerful greenhouse gas with a long atmospheric lifetime (3200 years). The validation of SF<sub>6</sub> emissions reported by developed countries has been a source for debate, again recently by Levin *et al.* [2010]. Better quantification of emissions from developing countries such as China will be especially important in better understanding the global budget of SF<sub>6</sub>.

#### 4. Summary

[20] The present study supports China's important role in the global atmospheric emissions of almost all major anthropogenic halogenated species, with global fractions reaching 20% for many individual species, and 19(14–27)% and 20(15–26)% for total global emissions of halogenated compounds when evaluated in terms of ODP and GWP (100-year time horizon), respectively. Along with signs of decreasing emissions of primary ODS, we also confirm the substantial increase in Chinese emissions of interim replacement HCFCs. China's emissions of many HFCs were also found to be significant fractions of the global budgets. Evidence suggests that most of these emissions are not driven by domestic Chinese consumption, the exception being HFC-134a emissions from mobile air conditioning. Emissions of these non-consumed HFCs should be better quantified in future bottom-up studies, the source of which could be large fugitive leaks during production. The emissions of PFCs and SF<sub>6</sub> in this study compare well with other estimates except for C<sub>3</sub>F<sub>8</sub>, which we find to have been underestimated. Uncertainties in our study arise from limitations of the correlation method, from limited spatial and temporal (especially in summer) sampling of Chinese emissions at Gosan, and from uncertainties in Chinese emissions of the tracer compound HCFC-22.

[21] **Acknowledgments.** We thank K.-S. Kang for help in operations at Gosan. We also thank AGAGE for sharing its instrumental expertise, calibration, and data processing capabilities, supported by NASA's Upper Atmosphere Research Program. Funding was provided from grant R01-2004-000-10844-0 (KOSEF) and the BK21 program at SEES, SNU. A. Stohl was supported by the Norwegian Research Council in the framework of the SOGG-EA project. Jooil Kim and Shanlan Li contributed equally to this work.

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- C. M. Harth, J. Mühle, P. K. Salameh, and R. F. Weiss, Scripps Institution of Oceanography, University of California, San Diego, 9500 Gilman Dr., La Jolla, CA 92093, USA.
- D.-J. Kang, Korea Ocean Research and Development Institute, Ansan PO Box 29, Seoul 426-744, South Korea.
- J. Kim, K.-R. Kim, S.-K. Kim, S. Li, and M.-K. Park, School of Earth and Environmental Sciences, Seoul National University, Seoul 151-747, South Korea. (krkim@snu.ac.kr)
- G. Lee, Environmental Science Division, Hankuk University of Foreign Studies, 89 Wangsan-Ri Mohyun-Myon Yongin-Shi Kyongki-Do, Yongin, Gyeonggido 449-741, South Korea.
- A. Stohl, Norwegian Institute for Air Research, PO Box 100, N-2027 Kjeller, Norway.