

Emissions of ozone-depleting halocarbons from China

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[1] National emission inventories of ozone-depleting substances (ODS) play a key role in the control mechanisms of the Montreal Protocol's emission reduction plans. New quasi-continuous ground-based atmospheric measurements allow us to estimate China's current emissions of the most effective ODS. This serves as an independent validation of China's ODS consumption data reported to the United Nations Environment Programme (UNEP). Emissions of most first-generation ODS have declined in recent years, suggesting compliance with the regulations of China's advanced phase-out program. In contrast the emissions of some second-generation ODS have increased. Because China is currently one of the largest consumers of first generation ODS, the country's upcoming complete phase-out will be crucial for the rate of decline of atmospheric ODS hence the eventual recovery of the stratospheric ozone. **Citation:** Vollmer, M. K., et al. (2009), Emissions of ozone-depleting halocarbons from China, *Geophys. Res. Lett.*, 36, L15823, doi:10.1029/2009GL038659.

1. Introduction

[2] The total atmospheric burden of anthropogenic ozone-depleting substances (ODS) has been decreasing steadily since the late 1990s as a result of the phase-out program under the Montreal Protocol and its various amendments [Clerbaux and Cunnold, 2007; Montzka et al., 1999]. Usage of the first generation ODS, including chlorofluorocarbons (CFCs), halons, methyl chloroform (CH_3CCl_3), carbon tetrachloride (CCl_4), and methyl bromide (CH_3Br), is generally banned in the Montreal Protocol's non-Article-5 (mainly developed) countries since 1996, but for the Article-5 countries, including China, their use is generally still permitted until their phase-out schedule for the first generation ODS is completed with a total ban by 2010 (and by 2015 for CH_3CCl_3). China's reported consumption (= production + import – export – feedstock) of ODS is of global signifi-

cance as it has amounted to over one third of the global consumption for the 3-year period 2004 – 2006 (UNEP Ozone Secretariat, Production and consumption of ozone depleting substances under the Montreal Protocol, 1986–2004, data updates to 2007, available at <http://www.unep.org/ozone>). In addition to the regulatory framework of the Montreal Protocol, China has accelerated its ODS phase-out by a few years, with a complete ban from use in new equipment for various usage sectors by 2005 – 2006 (UNEP Multilateral Fund, Phase-out plans and project (as of April 2008), 2008, available at <http://www.multilateralfund.org/files/Policy55Plans.pdf>).

[3] While consumption data are convenient regulatory quantities for ODS, emissions are more relevant to estimate the halogen loading to the atmosphere. Emissions differ from consumption because of the delayed escape of ODS from banks, i.e. reservoir ODS installed in equipment such as foams or refrigerators. Emissions can be derived from industry and government production and consumption data (Production, sales and atmospheric release of fluorocarbons through 2005, Alternative Fluorocarbons Environmental Acceptability Study, 2008, <http://www.afeas.org>, and UNEP Ozone Secretariat, Production and consumption of ozone depleting substances under the Montreal Protocol, 1986–2004, <http://www.unep.org/ozone>) combined with time-dependent emission functions, which account for the delayed releases [McCulloch et al., 2006; Midgley and Fisher, 1993]. For China, such 'bottom-up' emission data are still lacking.

[4] Regional emissions can also be derived from atmospheric observations ('top-down'), but for the Asian regions this method is limited to a few short-term studies [Blake et al., 2003; Palmer et al., 2003; Yokouchi et al., 2006]. Here we estimate for the first time Chinese emissions by inverse modeling of the first quasi-continuous high-precision measurements taken from October 2006 to March 2008, at the World Meteorological Organization's regional Global Atmosphere Watch station Shangdianzi in northern China. Our analysis includes the long-lived CFCs CFC-11 (CCl_3F), CFC-12 (CCl_2F_2), and CFC-113 ($\text{CCl}_2\text{FCClF}_2$), and CCl_4 , CH_3CCl_3 , chloroform (CHCl_3), the halons H-1211 (CBrClF_2) and H-1301 (CBrF_3), methyl bromide (CH_3Br), the hydrochlorofluorocarbons (HCFCs) HCFC-22 (CHClF_2) and HCFC-142b (CH_3CClF_2), and the greenhouse gas sulfur hexafluoride (SF_6).

2. Experimental Data

2.1. Sampling and Analysis

[5] Shangdianzi ($40^\circ 39.52'\text{N}$, $117^\circ 6.70'\text{E}$) is located at 280 m a.s.l. in a remote mountainous area approximately 100 km north of Beijing in the North China Plain. Our air trajectory analysis shows that this site is influenced by air

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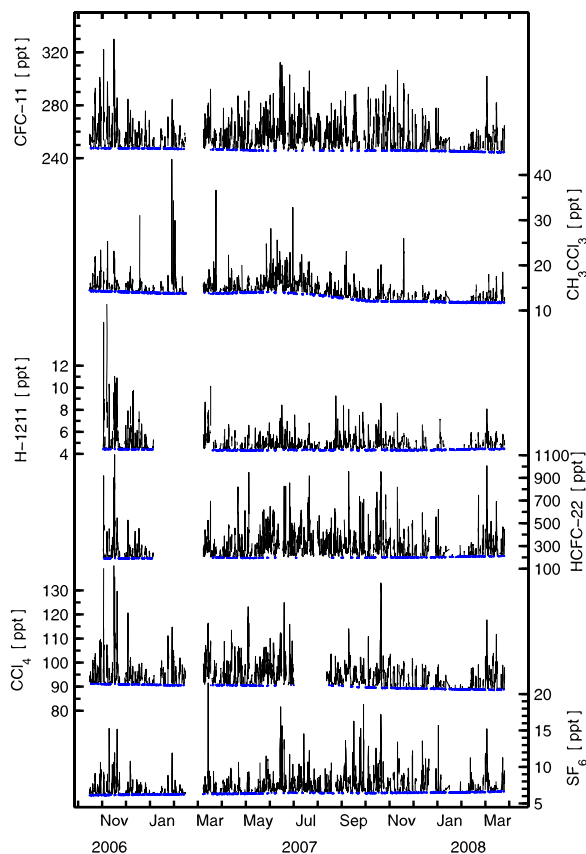


Figure 1. Ground-based measurements of selected halocarbons and sulfur hexafluoride (SF_6) at the Shangdianzi observatory, China. Mixing ratios are given as dry-air mole fraction in parts-per-trillion (ppt). Blue symbols denote measurements which were classified as regionally unpolluted.

masses that advect over some of the most densely-populated areas in the North China Plains carrying halogen pollution to the site, and occasionally (particularly during summer) by air masses from the south of China. Often air masses also arrive via the sparsely-populated regions of Siberia, Mongolia, and the Chinese province Inner Mongolia, carrying regionally unpolluted air to the site with background halogen mixing ratios typical for the northern hemisphere [Prinn *et al.*, 2000] (updated with AGAGE unpublished data per R. G. Prinn, personal communication, 2009).

[6] Air samples are analyzed in situ using a 2-channel electron-capture detector (ECD) gas chromatography (GC) technique coupled with a custom-built sample preparation system (see auxiliary material).¹ On the first channel, a packed molecular sieve 5A column is used in a first unit allowing for the rapid separation and detection of SF_6 and CFC-12 [Vollmer and Weiss, 2002] from a 2 ml sample loop, followed by separation from a second unit (10 ml sample loop, Silicone SP-2100 columns) for the subsequent analysis of CFC-11, CFC-113, CCl_4 , CH_3CCl_3 , CHCl_3 , C_2HCl_3 , and C_2Cl_4 [Prinn *et al.*, 2000]. The accuracies for the results of these substances are estimated at <5%. The second channel is equipped with a pre-concentration unit to

collect 500 ml sample, and with capillary columns (Poraplot-Q) and an oxygen-doped ECD for the analysis of H-1211, H-1301, CH_3Br , HCFC-22 and HCFC-142b [Miller, 1998]. The accuracies for this channel are estimated at <10%. Air samples and reference standards are alternately analyzed every 40 minutes. All results are reported on the Scripps Institution of Oceanography SIO-2005 primary calibration scale [Prinn *et al.*, 2000]. Time series for a selection of compounds are shown in Figure 1.

2.2. Inverse Method

[7] In our inverse modeling approach we combine our observations with 5-days backward calculations of the Lagrangian Particle Dispersion Model FLEXPART [Stohl *et al.*, 2005] run on the European Centre for Medium-range Weather Forecast wind fields using 6-hourly analysis fields (T799L91) interlaced with 6-hourly forecast fields (T + 3) yielding 3-hourly fields of $1^\circ \times 1^\circ$ horizontal resolution and 91 vertical levels (13 below 1000 m and 26 below 5000 m above model ground). From the obtained source-receptor relationships and the observed mixing ratios, *a posteriori* emissions distributions for the north-eastern part of China were estimated using an analytical inversion technique, which uses *a priori* emission distributions for regularization of the ill-posed inversion problem [Eckhardt *et al.*, 2008; Stohl *et al.*, 2009]. Total *a priori* emission estimates were horizontally distributed according to population density [Center for International Earth Science Information Network (CIESIN), 2005]. The inversion grid consists of grid boxes with different horizontal resolution to assure boxes of similar total residence time and therefore similar influence on the inversion result. The smallest gridbox was 0.5° by 0.5° . This leaves one grid box between the boxes occupied by Beijing and Shangdianzi. The resulting grid covers north-eastern China, Mongolia and parts of southern Siberia and North Korea (Figure 2). The total Chinese population within the inversion domain is estimated at 510 – 570 million (40% – 45% of the total population) according to population density distributions [CIESIN, 2005]. Assuming the same *per-capita* emissions for the Chinese regions not covered by our model, we scale our results to the total Chinese population. This up-scaling assumption was tested for CO using a CO inventory for China [Streets *et al.*, 2006] and found to add comparably little uncertainty to the overall emission estimates, and we assumed that this is also the case when applying this procedure to the ODS. While our technique currently does not include adequate error statistics for the uncertainty of the *a posteriori* emissions, these were explored by using an *a priori* emission range and related uncertainty ranges (see auxiliary material).

3. Results

[8] Our results show that China's emissions of most first-generation ODS (CFCs, halons, CH_3CCl_3 , CCl_4) have declined compared to those for 2001 based on short-term aircraft observations (Table 1), indicating that in general, the country is following the advanced phase-out plans (see <http://www.multilateralfund.org/files/Policy55Plans.pdf>). The emissions of the industrial solvents with short banking times, CH_3CCl_3 and CCl_4 , have declined since 2001 by ~70% and ~20% respectively. Over the same time, the

¹Auxiliary materials are available in the HTML. doi:10.1029/2009GL038659.

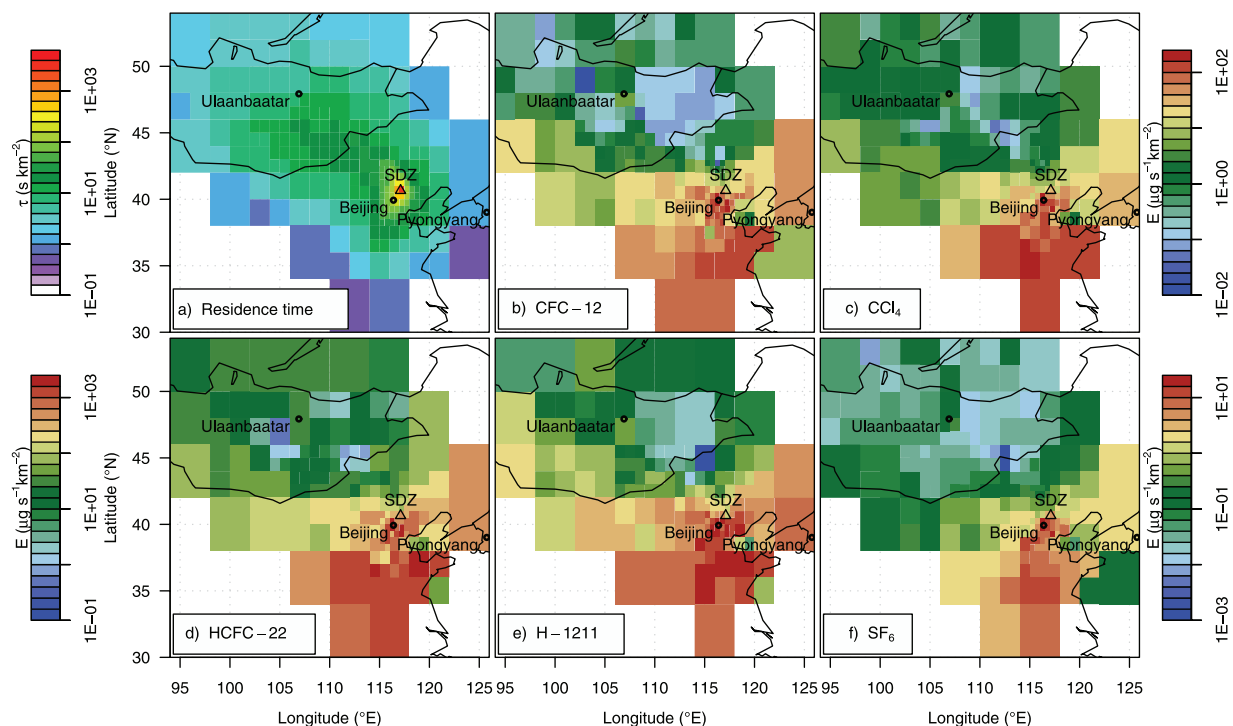


Figure 2. (a) Footprint for CFC-12 (log scale) for air masses reaching Shangdianzi (SDZ) Oct 2006 – March 2008 and (b–f) emission inventory maps for selected compounds (log scales). The open triangles in each map denote the location of Shangdianzi. Solid lines are borders to the ocean (right) and between China, Mongolia, and Russia to the north.

emissions of the refrigerant CFC-12 have declined by $\sim 55\%$ to 14 ($9 - 19$) kt a^{-1} . By contrast, we find that emissions of CFC-11 have increased by $\sim 40\%$ since 2001. This suggests that large amounts of CFC-11 were built into banks since 2001 and/or are potentially still used.

[9] For the 3 main CFCs, our aggregated emissions amount to 48 kt ODP a^{-1} (kt of the substances multiplied by their ozone depletion potential), ~ 8 times the consumption data reported to the United Nations Environment Programme (UNEP, $5.8 \text{ kt ODP a}^{-1}$) for 2007 (<http://www.unep.org/ozone>). This discrepancy is likely mainly due to the delayed emissions of banked CFC-11 from rigid foams and of CFC-12 leakage from installed refrigeration systems. For the solvent CFC-113 there is no basis for comparison, but its contribution is minor (0.8 kt a^{-1}) and is expected to vanish over the next few years due to its short banking time and its advanced ban from usage as a solvent (2006) and as a process agent (2008) (see <http://www.multilateralfund.org/files/Policy55Plans.pdf>). In contrast to the general decline in the emissions of the first-generation ODS, we find significant emissions for the second-generation ODS (HCFCs). The largest emissions for all our measured halocarbons are those for HCFC-22 (165 kt a^{-1}), a compound that is mainly used in stationary air conditioners. Our estimates substantially exceed those for 2004 ($52 \pm 32 \text{ kt}$) from observations at a station downwind of China [Yokouchi *et al.*, 2006] and indicate that consumption of this compound has increased strongly in recent years.

4. Global Perspective

[10] To put China's emissions in a global perspective we calculate worldwide emissions to the atmosphere, which we

infer from trends in atmospheric background mixing ratios (global 'top-down estimate') as measured by the AGAGE (Advanced Global Atmospheric Gases Experiment) global background network [Prinn *et al.*, 2000; Velders *et al.*, 2005]. Using atmospheric halocarbon observations from

Table 1. Chinese and Global Emissions of Ozone-Depleting Substances and Sulfur Hexafluoride to the Atmosphere^a

	China Emissions for 2001 ^b (kt a ⁻¹)	China Emissions, This Study ^c (kt a ⁻¹)	Global Top-Down Emissions for 2007 ^d (kt a ⁻¹)
CFC-11	23 ± 6	33 (26 – 43)	82 ± 30
CFC-12	30 ± 7	14 (9 – 19)	79 ± 40
CFC-113	–	0.8 (0.4 – 1.7)	5 ± 4
CH_3CCl_3	10.1 ± 2.6	3.3 (3.1 – 4.0)	–
CCl_4	18 ± 4.5	15 (10 – 22)	53 ± 30
H-1211	$2.5^e \pm 3.0$	2.1 (1.7 – 2.5)	6.2 ± 3.0
H-1301	–	0.09 (0.07 – 0.11)	2.3 ± 1.1
CH_3Br	2.9	0.24 (0.17 – 0.47)	–
HCFC-22	$51^f \pm 32$	165 (140 – 213)	365 ± 70
HCFC-142b	–	12 (10 – 18)	41 ± 5.0
HCFC-141b	–	–	60 ± 10
CHCl_3	–	86 (51 – 140)	–
SF_6	–	0.80 (0.53 – 1.10)	$7.5^g \pm 1.5$

^aThis study's mean results for China are derived using an uncertainty of 100% on the mean *a priori* values used in the inversion technique. The range's lower value of a compound was derived using the minimum of the lower *a priori* value and the 50% uncertainty, and the upper value by using the maxima of the upper *a priori* value and the 200% uncertainty.

^bBlake *et al.* [2003] and Palmer *et al.* [2003].

^cOct 2006 – March 2008.

^dGlobal estimates are based on global atmospheric observations. They are calculated in this study for 2007 based on methods described by Clerbaux and Cunnold [2007], Prinn *et al.*, [2000], and Velders *et al.* [2005].

^eSouth China only.

^fEstimate for 2004 [Yokouchi *et al.*, 2006].

^gEstimated by Velders *et al.* [2005].

several networks, growing discrepancies between global bottom-up and global top-down emission estimates have recently become apparent (UNEP Task Force on Emissions Discrepancies report, 2006, available at <http://www.unep.org/ozone>). These point to gaps in the understanding of the global budgets and may point to deficiencies in the emission estimates due to poorly known release rates from banks, or to inaccurate atmospheric lifetimes, due to some unknown or wrongly quantified removal processes from the atmosphere [Clerbaux and Cunnold, 2007]. Regional emission estimates can potentially help to address some of these discrepancies.

[11] The Chinese CFC-11 emissions account for more than one third of the global emissions but for CFC-12 this contribution is much smaller (18%). Here the CFC-12/CFC-11 emission ratio is <1 , opposite to that observed in recent years for other regions [Clerbaux and Cunnold, 2007] and may be a first indication from regional observations of a faster depleted bank for CFC-12 than for CFC-11. This reversed ratio is also suggested from the trend in global top-down emissions [Velders *et al.*, 2005]. By contrast, China's CFC-113 emissions are comparably small on a global scale, although the latter are associated with large uncertainties. Our results for CCl_4 can explain one third of the global top-down estimate (53 ± 30 kt) for 2007. Because emissions of CCl_4 from developed countries are expected to be insignificant due to the ban in 1996 and the short banking time, our results are indicative of some major other sources, possibly in other Article-5 countries, or other deficiencies in our understanding of its atmospheric budget. Some recent studies suggest reducing the CCl_4 atmospheric lifetime from 26 yrs to 20 yrs, however this would increase the gap between known sources and those inferred from global atmospheric background observations [Clerbaux and Cunnold, 2007].

[12] While its current emissions are of little relevance for ozone destruction, CH_3CCl_3 has been receiving special attention, because of its key role in inverse model calculations of the tropospheric 'cleansing agent' hydroxyl radical (OH) trends [Montzka *et al.*, 2000; Prinn *et al.*, 2005]. According to UNEP (see <http://www.unep.org/ozone>), global CH_3CCl_3 production has drastically declined in the last decade to a mean of 5.9 kt a^{-1} during 2005 – 2007 (66% Japan, 18% USA, and 13% China) and is accurately balanced by global consumption shared by mainly South Korea (50%) and China (40%) (see <http://www.unep.org/ozone>). Because of its short banking time of <2 yrs, this suggests that these 2 countries are currently the largest emitters of CH_3CCl_3 . Our calculated Chinese emissions of 3.3 kt a^{-1} are roughly in line with the country's consumption of 2.5 kt for 2007. This consumption is expected to vanish by 2010 when China's advanced phase-out plan targets a complete ban of this compound, 5 years ahead of the original Montreal Protocol agreements.

[13] The halons H-1211 and H-1301 are long-lived brominated halogens with large ozone-depleting potential (3 and 10 times that of CFC-11 respectively) and long atmospheric lifetimes (16 yrs and 65 yrs, respectively), which were predominantly used in mobile (H-1211) and stationary (H-1301) fire extinguishers. China has agreed on an accelerated phase-out plan (see <http://www.multilateralfund.org/files/Policy55Plans.pdf>) that encompasses a zero consumption

of H-1211 by 2006 and a capped consumption of 0.1 kt a^{-1} for H-1301 starting in 2006 until 2010 when this substance is also phased out completely. The discrepancy between the target consumption and our emission estimate of 2.1 kt a^{-1} for H-1211 is presumably due to the release from banks. These emissions contribute to one third of the global top-down emissions of 6.2 ± 3.0 kt a^{-1} . In contrast, Chinese H-1301 emissions (0.09 kt a^{-1}) contribute $<4\%$ to the global estimate (2.3 ± 1.1 kt a^{-1}). Our results may reflect the historically much smaller H-1301/H-1211 production ratio in the Article-5 countries compared to the non-Article-5 countries in Europe [Fraser *et al.*, 1999] where a historically higher production ratio is reflected in an equivalent higher observed emission ratio [Velders *et al.*, 2005].

[14] HCFC-22 and HCFC-142b emissions from China amount to nearly 45% and 30% of the global emissions for 2007, respectively. This is likely a consequence of rapidly growing production and consumption in China in the late 1990s and early 2000s [McCulloch *et al.*, 2006], which may have continued until present. However this is not clearly known, as recent data are only available from UNEP and in the form of an aggregated HCFC class (see <http://www.unep.org/ozone>). Our aggregated HCFC emissions (9.9 ODP kt) are currently smaller than the consumption of HCFCs in China (17.9 ODP kt) although we exclude HCFC-141b and HCFC-124 in the emission estimates. This points to a growing bank of these second-generation ODS in China, thus they will remain a persistent strong halogen source to the atmosphere for several more years. In addition, despite an accelerated phase-out plan, consumption is not officially required to be frozen until 2015 (UNEP Ozone Secretariat, 2007 Montreal adjustment on production and consumption of HCFCs, 2008 available at http://ozone.unep.org/Ratification_status/2007_Montreal_adjustments_on_hcfc.shtml).

[15] Our analysis also includes the non-ODS SF_6 , a very stable compound and potent greenhouse gas which is predominantly used in the high-voltage electric industry. Our estimate of 0.80 kt a^{-1} exceeds that based on bottom-up estimates for 2000 (0.36 kt a^{-1} [Olivier *et al.*, 2005]), and amounts to $\sim 11\%$ of the global emissions required (accurate top-down estimates are currently lacking) to account for the current atmospheric burden [Velders *et al.*, 2005]. Assuming that SF_6 emissions are proportional to a country's electrical power consumption, China's emissions would amount to $\sim 16\%$ of the global emissions in 2007 [Energy Information Administration, 2008], which is in reasonable agreement with our estimate based on the observations.

5. Conclusions

[16] Globally, the production and consumption of the first generation ODS will generally be completely phased out by 2010 (and by 2015 for CH_3CCl_3). It is hoped that a better quantification of the releases from the remaining banks will then result in a more accurately quantified rate of decline of the effective chlorine burden in the atmosphere and hence contribute to a better understanding of the recovery rate of stratospheric ozone. China's emissions of our measured substances total 82 ODP kt for 2007. Over the course of these upcoming years, this country's ODS emissions will

remain globally relevant and their monitoring therefore needs to be continued. A denser Asian network of atmospheric trace gas observations would improve spatial resolution, and could include other potent Asian emission regions of ODS and greenhouse gases that are relevant in a climate context.

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