Supplement of

A rise in HFC-23 emissions from eastern Asia since 2015

Hyeri Park et al.

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Figure S1: The sensitivity of the measurements to emissions of HFC-23 (a) for 2008–2019 and (b) for each year between 2008–2019. The figure (b) shows that the mean sensitivity of the observations to emissions from the eastern Asia region did not change substantially throughout this period. The red circles indicate locations of known HCFC-22 production plants.
HCFC-22 production in China

Note that under the Montreal Protocol, the use of HCFC-22 as a feedstock has been exempted from the phase-out schedule in some countries, including China. According to the TEAP 2021 report, the proportion of China’s HCFC-22 production for feedstock use has been increasing relative to HCFC-22 production for dispersive use (Figure S2). Therefore, the lack of a clear decline in HCFC-22 production even after 2013 (Figure S3) could be due to an increase in production for feedstock use in China.

Figure S2: Total HCFC-22 productions of China (stacked bars). Blue segments denote production for feedstock uses and purple for export.

Figure S3: (a) Total HCFC-22 productions of China (light gray bars) and HCFC-22 productions of eastern China in 2015 and 2018 (dark gray bars) (TEAP, 2021). (b) Inferred annual fractions of eastern China HCFC-22 productions to Chinese total productions (dark gray rhombi), extrapolated eastern China HCFC-22 production fractions for other years (green dashed line).
Figure S4: HFC-23 emissions estimates in eastern China (red line), in comparison with previous top-down estimates of Chinese HFC-23 emissions.
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**Table S1: Information on HCFC-22 production factories in China**
Observations of atmospheric HCFC-22 and estimation of top-down emissions in eastern China

High-frequency observation data of HCFC-22 at Gosan for 2008-2018 (Figure S6) show persistent pollution events, clearly implying that HCFC-22 emissions have been emanating from the surrounding regions, while the regional baseline concentrations of HCFC-22 show a similar increasing trend as the global NH baseline (green dots in Figure S6 for the Mace Head station). It should be noted that HCFC-22 baseline concentrations at Gosan drop periodically in summer due to strong intrusion of SH tropical air masses with low HCFC-22 concentrations during the East Asia summer monsoon (Li et al., 2018).

To confirm the link of HFC-23 emissions in eastern China to HCFC-22 production, we estimated HCFC-22 emissions from eastern China using the same inverse framework as for HFC-23 (Figure S7(a)). The continuing rise in the emissions seems to indicate that the contribution of dispersive use is still significant, although its production for dispersive use is currently being phased out in developing countries by the Montreal Protocol. HCFC-22 emissions from the whole of China were inferred from the faction of the population in eastern China. Results are consistent not only with previous studies but also with the inventory-based HCFC-22 emissions as shown in Figure S7(b), suggesting that population density still serves as good proxy for HCFC-22 emissions, and that the bottom-up emissions for the whole of China are relatively well-defined.
Figure S6: (a) Eastern Chinese emissions of HCFC-22 (red circles) derived from the atmospheric observations at Gosan, (b) Our HCFC-22 emission estimates by the whole of China (pink circles), determined by scaling up the eastern Chinese emissions by the fraction of the population (35%) that reside in eastern China (Rigby et al., 2019). Top-down Chinese emissions suggested in previous studies and bottom-up estimates are denoted by green rhombi and purple lines, respectively.
Three different spatial distributions of a priori emissions

Our inverse modelling results represent the mean and 2-σ uncertainty of 27 different model runs, where each set of three different a priori distributions (Figure S8) have 9 combinations of different a priori emission magnitudes. The first priori distribution is the “Population” a priori, determined based on the 2010 World population distribution (Warszawski et al., 2017). Population distribution has often been used as a reasonable first approximation when more specific information is not available (Stohl et al., 2010; Fang et al., 2019). The second priori distribution is the “Asiaflat” a priori, where the emissions within each country (whole of China, North Korea, South Korea, and whole of Japan) were evenly spread (Rigby et al., 2019; Park et al., 2021). This flattening may cause large a priori emissions to be allocated to western China and eastern Japan, where transport sensitivity was relatively low, while at the same time significantly lowering a priori emissions in eastern China and western Japan compared to other a priori distributions (Kim et al., 2021). The third priori distribution is the “EastAsiaflat” a priori, where the population distribution-based emissions are regionally flattened. Flatting regions are determined as the high sensitivity in the model domain (most of the high sensitivity area has less than or equal to 1° by 1°), i.e., eastern China, South Korea, North Korea, and western Japan. The region denoted “eastern China” contains nine provinces (Anhui, Beijing, Hebei, Jiangsu, Liaoning, Shandong, Shanghai, Tianjin, and Zhejiang) and “western Japan” contains four regions (Chūgoku, Kansai, Kyūshū & Okinawa and Shikoku) (Rigby et al., 2019; Park et al., 2021; Kim et al., 2021). “EastAsiaflat” a priori can be unbiased in terms of emission locations, such that the distribution of emissions in the posterior could point to likely emission hot spots, but such inference is reasonable only in regions where the influence of the observations is relatively strong. The a priori emissions were kept constants for all years, based on the 2008 emissions. For each a priori distribution, we tested 9 different combinations of a priori emission magnitudes and uncertainties, which are summarized in Table S2.

Note that our annual HFC-23 results represent the mean of 18 results from two different model runs a priori excluding “Asiaflat” a priori, because many HCFC-22 factories are located in eastern China, and thus HFC-23 emissions estimates for eastern China using “Asiaflat” a priori could be underestimated.
Figure S7: Three different spatial distributions of a priori used in this study. (a) a priori emissions map based on populations distribution. (b) flat emissions for each country. (c) flat emissions only in the regions with high sensitivity (eastern China, North Korea, South Korea and western Japan).
Table S2: List of 9 *a priori* configurations, with corresponding errors, applied to each *a priori* distribution. For each *a priori* configuration, the initial *a priori* estimate is multiplied by the listed scaling factor.

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Model validation based on CFC-11 emissions estimate from eastern China

To validate the optimization of our inversion framework, we analyzed CFC-11 emissions from eastern China using Gosan CFC-11 concentration data for 2008–2019. Top-down emissions of CFC-11 in East Asia were well-defined in a recent study (Park et al., 2021) which used multiple inversion methods.

Our results (Figure S9) showed good convergence among the runs for the same a priori distribution set, but relatively large (not statistically significant) difference between different a priori distributions, suggesting that a priori distributions have an impact on the a posteriori, and thus uncertainties associated with different a priori settings need to be considered to derive the full posteriori uncertainties.

Figure S10 shows that our CFC-11 emission estimates for eastern China are consistent within uncertainties with previously reported results from four different inverse methods that used Gosan observation data (only) (Park et al., 2021). Since the previous study with four different inverse models had used “Asiaflat” a priori, our estimation was also made with the same priori for direct comparison.

We took the mean of the a posteriori annual inversion results from 27 independent inversions with different a priori settings (see Table S2) as our final estimates of CFC-11 emissions from eastern China for 2008–2019 (Figure S11 and Table S3) and their uncertainties were defined as 2-σ of the resulting a posteriori emissions (95 % uncertainty), because the uncertainty of each inversion run can be considered fully correlated with each other. Our final emissions estimates of CFC-11 for eastern China are also consistent with previously reported results from four different inverse methods within uncertainties (Figure S11).

![Figure S8: CFC-11 emissions from eastern China derived using three different a priori distributions: “Population”, “Asiaflat”, “EastAsiaflat” a prioris. Each line represents the annual mean of 9 different model set-ups for each a priori distribution. Shading denotes 2-σ uncertainties.](image-url)
Figure S9: CFC-11 emissions from eastern China derived using the “Asiaflat” a priori distribution. Top-down emissions using FLEXPART-KNU (brown line) are compared to previously reported emissions from four different inverse methods using the same Gosan data for 2008–2019: NAME-HB (yellow crosses), NAME-InTEM (blue squares), FLEXPART-MIT (pink circles) and FLEXPART-Empa (gray triangles).

Figure S10: CFC-11 emissions estimate for eastern China derived in this study (FLEXPART-KNU, red circles) compared to previously derived emissions. Our annual results represent the mean of 27 different model runs, where each set of three different a priori distributions have 9 combinations of different emission magnitudes and 2-σ uncertainties (shading).
Table S3: Top-down emissions (Gg yr$^{-1}$) of CFC-11 and HCFC-22 for eastern China

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Simmonds, P. G., Rigby, M., McCulloch, A., Vollmer, M. K., Henne, S., Mühle, J., O'Doherty, S., Manning, A. J., Krummel, P. B., and Fraser, P. J.: Recent increases in the atmospheric growth rate and emissions of HFC-23 (CHF₃) and the link to HCFC-22 (CHClF₂) production, 2018.


Yao, B., Vollmer, M., Zhou, L., Henne, S., Reimann, S., Li, P., Wenger, A., and Hill, M.: In-situ measurements of atmospheric hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) at the Shangdianzi regional background station, China, Atmospheric Chemistry and Physics, 12, 10181-10193, 2012.