**China's hydrofluorocarbons (HFCs) emissions for 2011–2017
inferred from atmospheric measurements**

Bo Yao[†], Xuekun Fang^{*‡}, Martin K. Vollmer[∇], Stefan Reimann[∇], Liqu Chen[†],
Shuangxi Fang[†], Ronald G. Prinn[‡]

[†]Meteorological Observation Center of China Meteorological Administration (MOC/CMA),
Beijing 100081, China

[‡]Center for Global Change Science, Massachusetts Institute of Technology, Cambridge,
Massachusetts 02139, United States

[∇]Laboratory for Air Pollution and Environmental Technology, Empa, Swiss Federal Laboratories
for Materials Science and Technology, Dübendorf 8600, Switzerland

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11 ■ **TOC**

**Emissions of potent greenhouse gas
hydrofluorocarbons (HFCs)**

air conditioners and other sources

12

▪ ABSTRACT

Hydrofluorocarbons (HFCs) have been widely used in China to replace ozone-depleting substances (ODSs) that are required to be phased out under the Montreal Protocol regime. There are limited studies, which report HFC emissions in China, especially for recent years and using top-down approaches based on atmospheric measurements. Here we used flask and *in situ* measurements for nine HFCs from seven sites across China over the period 2011–2017, and FLEXPART-model-based Bayesian inverse modeling, to estimate HFC emission magnitudes and changes in China. We found that emissions of HFC-32 (CH_2F_2), HFC-125 (CHF_2CF_3), HFC-134a (CH_2FCF_3), HFC-227ea ($\text{CF}_3\text{CHFCF}_3$), and HFC-245fa ($\text{CHF}_2\text{CH}_2\text{CF}_3$) have been increasing fast over this period, while emissions of HFC-143a (CH_3CF_3), HFC-152a (CH_3CHF_2), HFC-236fa ($\text{CF}_3\text{CH}_2\text{CF}_3$), and HFC-365mfc ($\text{CH}_3\text{CF}_2\text{CH}_2\text{CF}_3$) were relatively stable. Total CO_2 -equivalent emissions of the nine HFCs increased from $\sim 60 \text{ Tg yr}^{-1}$ in 2011 to $\sim 100 \text{ Tg yr}^{-1}$ in 2017. Among these nine HFCs, HFC-134a (39%) and HFC-125 (35%) are the biggest contributors to the national total HFC CO_2 -equivalent emissions. Cumulative contributions from China's HFC emissions to the global total HFC mole fractions and their related radiative forcing increased from 1.0% in 2005 to 10.7% in 2017. When comparing global emissions with the sum of emissions from China and the developed countries, an increasing difference is observed over recent years, which points to substantial additional HFC emissions from other developing countries under the Kyoto Protocol.

■ INTRODUCTION

Hydrofluorocarbons (HFCs) have only a negligible effect on stratospheric ozone loss.^{1, 2} Thus, they have been used to replace ozone-depleting chlorofluorocarbons (CFCs), halons and hydrochlorofluorocarbons (HCFCs), in refrigeration, air conditioning, foam blowing and other applications since the 1990s. This is in compliance with the Montreal Protocol that was agreed on in 1987, to control consumption and production of ozone-depleting substances (ODSs).¹ However, most HFCs are potent greenhouse gases with high global warming potentials (GWPs).¹ The Kigali Amendment to the Montreal Protocol was agreed upon in 2016, and sets up schedules for limiting the HFC production and consumption in both developed and developing countries. Previous studies showed that atmospheric mole fractions of HFCs have been increasing globally between 2012–2016 by an average of 1.6 ppt yr⁻¹ (parts per trillion) for HFC-32 (CH₂F₂), 2.1 ppt yr⁻¹ for HFC-125 (CHF₂CF₃), 5.6 ppt ppt yr⁻¹ for HFC-134a (CH₂FCF₃), and 1.5 ppt yr⁻¹ for HFC-143a (CH₃CF₃), and these rates are faster than average increases reported for 2008 to 2012.^{1, 3}

Along with the phase-out process of CFCs and HCFCs in compliance with the Montreal Protocol, HFCs have become widely used as replacements in China and other parts of the world since the 1990s. For example, in new mobile air conditioners in China, HFC-134a has replaced CFC-12 (CCl₂F₂) as a refrigerant since around 2000.⁴ More recently HFCs are now widely used to replace HCFCs, whose production and consumption were frozen in 2013 and will be phased-out by 2030. For example, HCFC-22 (CHClF₂), which was the predominant refrigerant used in the room air-conditioning sector, is currently replaced by R-410A (a blend of HFC-32 and HFC-125).^{5, 6} Apart from the Kigali Amendment to the Montreal Protocol with a freeze on production

and consumption of HFCs in 2024, no additional regulations on HFCs (excluding HFC-23 (CHF₃)) are currently in force in China.

Bottom-up inventory-based emission estimates show that total Chinese HFC emissions increased rapidly from 35 Gg yr⁻¹ in 2005 to 76 Gg yr⁻¹ in 2013.⁶ No more inventory-based emission estimates are available beyond 2013. Projections suggest that China's HFC emissions would have increased substantially until 2050 if not limited as foreseen by the Kigali Amendment.^{6, 7}

HFC emissions from China have already been estimated in the past based on atmospheric measurements, using both ratio methods and inverse modeling methods.⁸⁻¹⁵ For example, atmospheric measurements of HFCs at the Gosan station (South Korea), combined with a tracer-ratio method, have been used to estimate HFC emissions in China for 2008.^{10, 11} Also, measurements of HFCs at Gosan, Hateruma (Japan), and Cape Ochi-ishi (Japan) have been used to estimate HFC emissions for 2005–2006¹², 2008¹³ and 2007–2012¹⁴ from East Asian countries using an inverse modeling approach. However, these studies only provide top-down estimates for certain years through 2012 for selective HFCs. Consequently, information on China's HFC emissions after 2013 is lacking. Also, so far no top-down emission estimates for HFC-227ea, HFC-245fa, and HFC-365mfc have been reported for China, although bottom-up emissions estimates are available for HFC-227ea and HFC-245fa for a limited period of 2005–2009.⁶

This study provides comprehensive emission estimates for nine HFCs (see information on nine HFCs in Table S1) in China using the atmospheric measurement data over 2011–2017 and explores the HFC changes in the context of a fast dynamic ODS phase-out process in China. The nine HFCs discussed here are HFC-32 (CH₂F₂), HFC-125 (CHF₂CF₃), HFC-134a (CH₂FCF₃), HFC-143a (CH₃CF₃), HFC-152a (CH₃CHF₂), HFC-227ea (CF₃CHF₂CF₃), HFC-236fa

(CF₃CH₂CF₃), HFC-245fa (CHF₂CH₂CF₃) and HFC-365mfc (CH₃CF₂CH₂CF₃). HFC-23 (CHF₃) was not included in this study, because it is a by-product from HCFC-22 (CHClF₂) production, while the nine HFCs in this study are from intentional uses.

■ MATERIALS AND METHODS

Sampling and Instrument Measurement

Detailed information for the sampling and the measurement of atmospheric HFCs is provided in a previous study.¹⁶ Here we give a brief overview. Seven Chinese flask sampling sites were used in this study (Figure 1): Heyuan (HYN; 23.69°N, 114.60°E), Shangri-La (XGL; 27.48°N, 99.00°E), Jiangjin (JGJ; 29.15°N, 106.15°E), Lin'an (LAN; 30.18°N, 119.44°E), Mount Waliguan (WLG; 36.29°N, 100.90°E), Shangdianzi (SDZ; 40.65°N, 117.12°E) and Longfengshan (LFS; 44.73°N, 127.60°E). Flask sampling started in September 2010 at SDZ, LAN, and LFS, in September 2010 at WLG, in July 2011 at XGL, in January 2017 at HYN, and in March 2017 at JGJ. The sampling frequency is daily for the HYN and JGJ sites and weekly for the other sites. At each site, ambient air was sampled in 3-L canisters, and then the canisters were shipped to the chemical analysis laboratory in Beijing. A custom-built “Medusa” gas chromatographic system with mass spectrometric detection (Agilent 6890/5975B, USA) was used to measure the mole fractions of HFCs.^{17, 18} The measured HFC mole fractions in this study are linked to Advanced Global Atmospheric Gases Experiment (AGAGE) reference standards and are reported as dry air mole fractions on the primary calibration scales developed at the Scripps Institution of Oceanography (SIO).^{17, 18} Note that in the years of 2011–2012 and 2015–2017 additional *in situ* measurements of atmospheric HFCs are available for the SDZ, using the same type of instrument and analysis procedure as in the flask analysis. The high-frequency (~2h) *in situ* data were averaged into daily resolution to reduce temporal correlations among

data. These additional *in situ* data help constrain HFC emissions in northern China. All flask data for each HFC from each site were filtered to remove some extreme outliers using the Tukey's fence approach. Only 241 out of 16704 (1.4%) flask data were flagged out. All flask and *in situ* observation data are shown in Figures S1-S9 for the nine HFCs, respectively.

Inverse Modeling of Emissions

The inverse modeling approach used in this study is a FLEXPART ("FLEXible PARTicle dispersion model")-based Bayesian inversion, which is adopted from an earlier study.¹⁹ FLEXPART is a Lagrangian transport and dispersion model that is used to simulate air parcel trajectories. It is suitable for the simulation of a large range of atmospheric transport processes.²⁰ ²¹ Driven by meteorological data (European Centre for Medium-Range Weather Forecasts - ECMWF) with 1° x 1° global resolution and 3-hourly temporal resolution, the FLEXPART model is run in a backward mode in time for 20 days to establish a source-receptor relationship matrix, hereafter called "emission sensitivities" (Figure S10 in Supplementary Information). The FLEXPART emission sensitivities were combined with the atmospheric HFC measurements and a Bayesian optimization technique to derive the emission strengths in grid boxes in China. The cost function is

$$J(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x}_a)^T S_a^{-1}(\mathbf{x} - \mathbf{x}_a) + \frac{1}{2}(\mathbf{y}^{\text{obs}} - H\mathbf{x}_a)^T S_o^{-1}(\mathbf{y}^{\text{obs}} - H\mathbf{x}_a).$$

We find the minimum by solving $\nabla_x J(x) = 0$, which yields

$$\mathbf{x} = \mathbf{x}_a + S_a H^T (H S_a H^T + S_o)^{-1} (\mathbf{y}^{\text{obs}} - H\mathbf{x}_a), \text{ and}$$

$$S_b = (H^T S_o^{-1} H + S_a^{-1})^{-1}.$$

121 Here $\mathbf{x}$ is the state vector of emission strength ($\text{g m}^{-2} \text{s}^{-1}$) in each grid cell, $\mathbf{x}_a$ is the prior emission
 122 vector, $\mathbf{H}$ is the source-receptor relationship matrix got from FLEXPART backward simulations,
 123 $\mathbf{S}_a$ is the prior emission error covariance matrix, $\mathbf{S}_b$ is the posterior emission error covariance
 124 matrix, $\mathbf{y}^{\text{obs}}$ is the HFC measurement vector, and $\mathbf{S}_o$ is the observational error covariance matrix.
 125 There is no gridded emission inventory available for all our HFCs. Thus, we used HFC global
 126 total emissions in 2011²² disaggregated approximately according to the population spatial
 127 distribution²³. The map of prior HFC-32 emissions as an example is shown in Figure S11. Our
 128 tests show that after scaling prior emissions by a factor of 2 total posterior emissions for China
 129 from inverse modeling only changed by few percentages (maximum of 13% for HFC-152a as
 130 tabulated in Table S2 for all nine HFCs). Prior emissions for a specific HFC were the same for
 131 all years during 2011–2017. There is no knowledge of prior emission uncertainties ($\mathbf{S}_a$). Here we
 132 set the prior emission uncertainty to be 500% of the emission in each grid box, squared values of
 133 which are the diagonal elements of $\mathbf{S}_a$. The observational error covariance matrix $\mathbf{S}_o$ consists of
 134 measurement precision error, representation error and background error. More information on
 135 constructing $\mathbf{S}_o$ and background is provided in the Supplementary Information. The resulting
 136 annual emissions of each HFC derived from this inverse modeling are shown in Table 1. The
 137 map of posterior HFC-32 emission as an example from inverse modeling is shown in Figure S12.
 138 Modeled and observed mole fractions at sampling sites for HFC-32 as an example are shown in
 139 Figure S13. Figures S14-S31 show 1:1 plots of observed and modeled mole fractions of each
 140 HFC (both plots with and without background mole fractions for each HFC). Relatively poor
 141 model simulations lead to relatively high posterior emission uncertainty.

Estimation of Chinese Contributions to the Global Atmospheric Mole Fractions and Radiative Forcing

The approach to estimate the atmospheric mole fractions related to Chinese emissions only is the same as used in previous studies.^{6, 7} Briefly, emissions for China derived from Fang et al. for 2005–2010⁶ and this study for 2011–2017 are used to calculate atmospheric mole fractions due to Chinese emissions of a specific HFC i in the year j , taking into account lifetime, molecular weight, etc. (see equations (1), (2) and (3)). Atmospheric radiative forcing for each HFC ($RF_{i,j}$) was then calculated using the mole fractions of each HFC multiplied by their radiative efficiency (RE_i ; listed in Table S1) (see equation (4)).

$$\frac{dC_i}{dt} = F_i \times E_i - \frac{C_i}{\tau_i} \quad (1)$$

$$C_{i,j} = C_{i,j-1} \times \exp\left(-\frac{1}{\tau_i}\right) + F_i \times E_{i,j-1} \times \tau_i \times (1 - \exp\left(-\frac{1}{\tau_i}\right)) \quad (2)$$

$$F_i = \left(\frac{N_A}{N_a}\right) \frac{F_{surf}}{M_i} = 5.68 \times 10^{-9} \frac{F_{surf}}{M_i} \quad (3)$$

$$RF_{i,j} = C_{i,j} \times RE_i \quad (4)$$

Here $E_{i,j-1}$ are the annual HFC emissions (kg yr^{-1}), $C_{i,j}$ and $C_{i,j-1}$ are the mean surface mole fractions (ppt), τ_i is the lifetime (yr), M_i is the HFC molecular weight (kg mol^{-1}), N_a is the number of molecules in the earth atmosphere (1.06×10^{32}), N_A is the Avogadro constant ($6.02 \times 10^{23} \text{ mol}^{-1}$), F_i (ppt kg^{-1}) is a factor that relates the mass emitted to the global mean surface mole fractions, and F_{surf} is a dimensionless factor relating the global mean surface mole fraction to the global mean atmospheric mole fraction. The factor 1.07 was used as F_{surf} for all HFCs.^{24, 25} τ_i and M_i are listed in Table S1.

■ RESULTS AND DISCUSSION

China's Emissions Derived from Inverse Modeling

Figure 2 shows increasing emissions for HFC-32, HFC-125, HFC-134a, HFC-227ea and HFC-245fa in China during 2011–2017. Over this period, HFC-32 emissions increased from 4.4 (3.8–5.0) Gg yr⁻¹ to 11.3 (10.5–12.0) Gg yr⁻¹ (Figure 2a) and HFC-125 emissions increased from 4.7 (3.5–5.9) Gg yr⁻¹ to 10.8 (9.7–11.9) Gg yr⁻¹ (Figure 2b). The concurrences of absolute magnitude and emission increase between HFC-32 and HFC-125 are consistent with the fact that R-410A (a blend of HFC-32 (50% by mass) and HFC-125 (50% by mass)) has been increasingly used as the refrigerant in new room air conditioners since around 2005.^{26, 27} HFC-134a emissions showed a rapid increase during 2011–2015, and then stabilized. HFC-227ea is mainly used as an extinguishing agent in the firefighting sector, and our results show that the emissions increased by four times over the period, reaching 1.4 (1.3–1.5) Gg yr⁻¹ in 2017. HFC-245fa (mainly used as a foam blowing agent) also increased quickly, from 0.75 (0.5–0.9) Gg yr⁻¹ in 2011 to 1.3 (1.2–1.5) Gg yr⁻¹ in 2017.

On the other hand, HFC-143a, HFC-152a, HFC-236fa, and HFC-365mfc emissions were relatively stable over the whole period, with ranges of 2.1–3.2 Gg yr⁻¹, 4.1–5.0 Gg yr⁻¹, 0.08–0.13 Gg yr⁻¹ and 0.34–0.42 Gg yr⁻¹, respectively (Figure 2). HFC-143a is mainly used as a refrigerant in the industrial/commercial refrigeration sector; HFC-152a is mainly used in the foam blowing sector and in the aerosol sector; HFC-236fa is mainly used as an extinguishing agent in specialized applications; and HFC-365mfc is mainly used as a foam blowing agent. An industrial report²⁸ shows that consumptions of HFC-143a, HFC-152a, and HFC-236fa during 2011–2015 (no data after 2015) did not increase, which may explain our derived stable emissions of these three HFCs. There were no atmospheric measurements for HFC-365mfc for

2011–2013, and the measurements of HFC-365mfc since 2014 inferred stable emissions. There is no information available for HFC-365mfc production, consumption and bottom-up emission estimates during 2011–2017 in China. Thus, this study provides the first HFC-365mfc emission estimates for China.

Comparisons to Other Studies

Comparisons of estimated emissions of the four major HFCs (HFC-32, HFC-125, HFC-134a, and HFC-143a) between this study and previous studies are shown in Figure 3. It shows overall good agreements for both magnitudes and temporal trends of HFC-125, HFC-134a, and HFC-143a emissions. As for HFC-32, our emission estimates agree well with both the CO-based regression analysis and the CO-based ratio analysis by Yao et al.¹⁵, while our estimates are at the lower end of the 7.0 (4.9–9.1) Gg yr⁻¹ averaged over 2010–2012 reported by Lunt et al.¹⁴. They are also lower than the bottom-up estimates (7.7 Gg yr⁻¹ in 2011 – 12.0 Gg yr⁻¹ in 2013) by Fang et al.⁶. Top-down estimates suggest that HFC-32 emissions increase at a smaller rate than what was proposed from bottom-up estimates by Fang et al.⁶. This in part suggests that the estimated consumption based on production or the emission factors used in the bottom-up inventory study might be too high, and that all influencing factors need more investigations in future studies. However, please note that HFC-32 estimates do not significantly influence the total HFC emission estimates since HFC-32 emissions only contributed to ~6% of total HFC CO₂-eq emissions (Figure 4). As for HFC-125, both top-down studies and a bottom-up study show a rapid increase in HFC-125 emissions during the 2008–2017 period (Figure 3b). Compared to other HFCs, a larger number of studies have estimated HFC-134a emissions (Figure 3c), which increased from ~5 Gg yr⁻¹ in 2005 to ~30 Gg yr⁻¹ in 2013. Apart from this current study, no other

studies have estimated HFC-134a emissions after 2013. As for HFC-143a emissions, the bottom-up study⁶ and this study shows an agreement in the magnitude of the emissions (Figure 3d).

CO₂-eq Emissions and Proportions among HFCs

Since HFCs are potent greenhouse gases, it is important to understand the relative contributions of specific HFCs and their evolutions in terms of CO₂-equivalent emissions. The HFC CO₂-eq emissions were obtained using the emissions of each HFC multiplied by their 100-year GWPs (GWP₁₀₀; see Table S1). The largest relative emissions, both by mass and CO₂-eq, are found for HFC-134a, being 48% and 39% on average over 2011–2017, respectively (Figure 4). HFC-125 contributed about ~18% to the total mass emissions, while it contributed about 35% to the total CO₂-eq emissions, due to its relative high GWP (GWP₁₀₀ = 3500) compared to the average GWP (GWP₁₀₀ = 2784) of the nine HFCs used in this study. Combined emissions of HFC-32, HFC-125, HFC-134a, and HFC-143a contributed 94% of all HFC CO₂-eq emissions over 2011–2017, while the other five HFCs contributed ~6%. Thus, these four major HFCs are a current and future focus.

The contributions of each substance to total CO₂-eq emissions from HFCs changed over 2011–2017 (Figure 4 d and e). For example, HFC-227ea emission contributions increased from 2.0% in 2011 to 4.4% in 2017, while those for HFC-134a decreased from 47% in 2011 to 34% in 2017. Among these nine HFCs, HFC-32, HFC-125, HFC-227ea, and HFC-245fa show increasing contributions to the total CO₂-eq emissions, which suggest their increasing importance among the HFCs.

Global and Other Regional Emissions

Aggregated HFC emissions reported by Annex I countries (industrialized countries and economies in transition) to the United Nations Framework Convention on Climate Change (UNFCCC) increased from 240 Tg CO₂-eq yr⁻¹ in 2005 to 361 Tg CO₂-eq yr⁻¹ in 2016, the latest year for which UNFCCC reporting is available (Figure 5a).²⁹ For comparison, global total HFC CO₂-eq emissions are used here based on emissions derived from observation at remote AGAGE stations.^{22, 30} These emissions increased from 320 Tg yr⁻¹ in 2005 to 739 Tg yr⁻¹ in 2016 (Figure 5a). Thus, the contribution from Annex I countries to global totals decreased from 75% in 2005 to 49% in 2016.

To estimate China's contribution, we adopted the emissions estimated for 2005–2010 by Fang et al.⁶ and emissions estimated for 2011–2017 by this study (Figure 5a). Contributions from aggregated Chinese HFC CO₂-eq emissions to the global HFC total CO₂-eq emissions increased from 2.6% in 2005 to 15% in 2016. AGAGE atmospheric measurements show that total mole fractions of the nine HFCs in the global atmosphere increased from 49 ppt (derived from http://agage.eas.gatech.edu/data_archive/global_mean/; equivalent to 8 mW m⁻² using the radiative efficiencies listed in Table S1) in 2005 to 168 ppt (28 mW m⁻²) in 2017 (Figure S32). Our modeled atmospheric mole fractions for China (see MATERIALS AND METHODS) show contributions of 0.08 mW m⁻² in 2005 and 3.0 mW m⁻² in 2017 (Figure S32). This means that cumulative contributions from China's HFC emissions to global total HFC radiative forcing increased from 1.0% (uncertainty was not estimated because no uncertainty information is available for the 2005–2010 inventory estimates) in 2005 to 10.7% in 2017 (Figure 5b). Thus, China's HFCs pose increasing importance to global HFC CO₂-eq emissions and future climate change mitigation.

For 2005–2016, the summed emissions from Annex I countries and China were lower than the global total HFC CO₂-eq emissions (Figure 5a). Importantly, this emission gap increased over 2005–2016, which suggests substantial increases in HFC use and emissions in developing countries (not obligated to report emissions to the UNFCCC) other than China. However we acknowledge that this statement is only correct, if observation-based global total HFC emission estimates, the reported emissions from Annex I countries to the UNFCCC and the top-down estimated emissions for China are accurate and don't have temporal biases. A recent study for India, based on a 2-month aircraft campaign,³¹ shows that HFCs emissions for this country were 38 (29–48) CO₂-eq Tg yr⁻¹ in 2016. These emissions account for a small fraction of the global gap of 268 CO₂-eq Tg yr⁻¹ in 2016 given the population size of India. Unfortunately, atmospheric HFC measurements are not available in many regions, e.g., Southeast Asia, Africa, South America, which could be significant contributors to the global HFC emissions. Thus, to better understand global and regional HFC emission patterns, it is encouraged to carry out atmospheric measurements of HFCs in areas that are likely to contribute significantly to global emissions, and carry out similar top-down emission estimates as in the present study.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: xxxxxx. Additional information including inversion setups, HFC information, average emission sensitivity, prior and posterior emissions, observed and modeled mole fraction time series, and radiative forcing. HFC mole fraction measurement data for the flask and *in situ* sites can be accessed by contacting Bo Yao (yaob@cma.gov.cn).

272 ▪ **AUTHOR INFORMATION**

273 **Corresponding Author**

274 *Phone: 617-955-9144. E-mail: fangxk@mit.edu; fangxuekun@gmail.com

275 **ORCID**

276 Xuekun Fang: 0000-0002-7055-0644

277 **Notes**

278 The authors declare no competing financial interest.

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398 ■ **TABLES**

399 **Table 1.** Emissions (Gg yr⁻¹; 1-sigma uncertainty) of each HFC from China derived from inverse modeling for 2011–2017. There are
 400 no emission estimates of HFC-365mfc for 2011–2013 since only *in situ* measurements of HFC-365mfc at the SDZ site are available
 401 for 2011–2013 and therefore the measurement data were not used to estimate HFC-365mfc emissions in China.

	HFC-32	HFC-125	HFC-134a	HFC-143a	HFC-152a	HFC-227ea	HFC-236fa	HFC-245fa	HFC-365mfc
2011	4.4 (3.8–5.0)	4.7 (3.5–5.9)	19 (17–22)	2.1 (1.3–2.8)	4.8 (4.1–5.4)	0.4 (0.3–0.5)	0.09 (0.06–0.12)	0.7 (0.5–0.9)	N/A
2012	4.7 (3.5–6.0)	6.0 (3.8–8.2)	15 (10–21)	2.2 (1.2–3.3)	4.1 (3.2–4.9)	0.5 (0.4–0.7)	0.12 (0.08–0.17)	0.7 (0.4–1.1)	N/A
2013	8.0 (6.1–10.0)	9.8 (7.1–12.6)	24 (15–33)	2.7 (1.8–3.6)	5.0 (3.5–6.4)	0.7 (0.4–0.9)	0.11 (0.07–0.16)	0.7 (0.4–1.0)	N/A
2014	7.3 (5.9–8.6)	9.0 (7.1–10.9)	27 (21–33)	2.5 (1.7–3.3)	4.1 (2.8–5.5)	1.0 (0.7–1.2)	0.08 (0.05–0.12)	0.8 (0.5–1.1)	0.3 (0.1–0.6)
2015	7.3 (6.1–8.5)	11.3 (8.0–14.7)	32 (24–41)	3.9 (2.5–5.3)	5.9 (3.7–8.0)	1.2 (0.9–1.5)	0.13 (0.08–0.18)	1.6 (1.1–2.1)	0.4 (0.1–0.7)
2016	10.1 (8.7–11.6)	11.2 (9.0–13.3)	30 (24–36)	3.2 (2.3–4.1)	5.0 (3.9–6.1)	1.1 (0.9–1.3)	0.10 (0.07–0.13)	1.2 (0.9–1.5)	0.4 (0.3–0.6)
2017	11.3 (10.5–12.0)	10.8 (9.7–11.9)	25 (22–27)	3.1 (2.6–3.6)	4.3 (3.8–4.8)	1.4 (1.3–1.5)	0.13 (0.12–0.14)	1.3 (1.2–1.5)	0.4 (0.3–0.4)

402

FIGURES

Figure 1. Sampling sites used in this study. The sites are Heyuan (HYN; 23.69°N, 114.60°E), Shangri-La (XGL; 27.48°N, 99.00°E), Jiangjin (JGJ; 29.15°N, 106.15°E), Lin'an (LAN; 30.18°N, 119.44°E), Mount Waliguan (WLG; 36.29°N, 100.90°E), Shangdianzi (SDZ; 40.65°N, 117.12°E) and Longfengshan (LFS; 44.73°N, 127.60°E). The figure and image is made with GeoMapApp (www.geomapapp.org) / CC BY / CC BY³² and the China boundary file is from the software MeteoInfo (<http://www.meteothink.org/index.html>).

Figure 2. Annual emissions of HFCs in China derived from inverse modeling using atmospheric observations at seven sites.

Figure 3. Estimates of HFC emissions in China during the period 2005–2017. The emission estimates are from top-down⁸⁻¹⁵ and bottom-up^{6, 33, 34} approaches. The x-axis error bar in the plot represents the span of the target period in the respective study, for example the 14 months from November 2007 to December 2008 in Li et al.¹¹ and the 3 years from 2010 to 2012 in Lunt et al.¹⁴

Figure 4. Proportions of each HFC to totals in China, in terms of mass and CO₂-eq emissions. The symbol * denotes the combined proportion of HFC-236fa, HFC-245fa, and HFC-365mfc to total HFCs.

425

426 **Figure 5.** Global HFC emissions and China's contributions to the global atmospheric HFC
 427 radiative forcing. CO₂-eq emissions for Annex I countries were derived from UNFCCC.²⁹ Global
 428 total CO₂-eq emissions were based on emissions derived from observations at remote AGAGE
 429 stations.^{22, 30} Emissions for China were derived from Fang et al. for 2005–2010⁶ and this study
 430 for 2011–2017. The “aggregated” error bars shown in the China's HFC CO₂-eq emissions
 431 during 2011–2017 are the sum of the posterior emission uncertainty multiplied by the
 432 corresponding GWP value for each HFC. Calculation of contributions from China's HFC
 433 emissions to global HFC radiative forcing used the AGAGE atmospheric measurements

434 (http://agage.eas.gatech.edu/data_archive/global_mean/). In the b panel, uncertainties were not
435 estimated because uncertainties were not available for China's HFC emissions during
436 2005–2010⁶.