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A decline in emissions of CFC-11 and related chemicals from eastern China

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Emissions of ozone-depleting substances, including trichlorofluoromethane (CFC-11), have decreased since the mid-1980s in response to the Montreal Protocol^{1,2}. Recently, an unexpected increase in CFC-11 emissions since 2013 was reported, with much of the global rise attributed to emissions from eastern China^{3,4}. Here we use high-frequency atmospheric mole fraction observations from Gosan, South Korea, and Hateruma, Japan, together with atmospheric chemical transport model simulations, to investigate regional CFC-11 emissions from eastern China. We find that CFC-11 emissions returned to pre-2013 levels in 2019 (5.0 ± 1.0 Gg yr⁻¹ in 2019 compared to 7.2 ± 1.5 Gg yr⁻¹ for 2008–2012, 1 s.d.), decreasing by 10 ± 3 Gg yr⁻¹ since 2014–2017. Furthermore, we find that in this region carbon tetrachloride and dichlorodifluoromethane (CFC-12) emissions, potentially associated with CFC-11 production,

were higher than expected after 2013 and then declined one to two years prior to the CFC-11 emissions reduction. This suggests CFC-11 production in eastern China after the mandated global phase-out, and a subsequent decline in production during 2017-2018. We estimate that the CFC-11 bank in eastern China is up to 112 Gg larger in 2019 compared to pre-2013, most likely as a result of recent production. Nevertheless, it seems that any substantial delay in ozone layer recovery has been avoided, perhaps due to timely reporting^{3,4} and subsequent action by industry and government in China^{5,6}.

An unexpected rise in global emissions of CFC-11 beginning in 2013 was detected by two independent global atmospheric measurement networks, operated by the National Oceanic and Atmospheric Administration (NOAA)³ and the Advanced Global Atmospheric Gases Experiment (AGAGE)^{4,7}. Data collected in the background atmosphere were used to infer a rise in global emissions of 17 ± 3 Gg yr⁻¹ (NOAA) or 11 ± 3 Gg yr⁻¹ (AGAGE) for 2014 - 2017, compared to 2008 - 2012⁴. The enhanced emissions to date are unlikely to lead to a substantial delay in ozone recovery, although a delay of years to decades would be expected if emissions continued at the levels observed between 2014 and 2017^{8,9}.

Under the Montreal Protocol, production and consumption of CFC-11 for emissive applications was phased-out globally in 2010. Before the phase-out, CFC-11 was used initially as a propellant gas in aerosol cans, as a refrigerant in chillers, and later as a blowing and insulating agent in the manufacture of foams in buildings and domestic appliances¹⁰. As a result, substantial “banks” of CFC-11 remain (i.e. CFC-11 contained in foams and chillers), and their emissions are expected to continue for several decades at a gradually declining rate¹¹. Largely because the observed emissions rise since 2013 was contrary to the expectation of globally and regionally falling emissions from banks, it was concluded that the rise since 2013 was indicative of new CFC-11 production, most likely for the manufacture of closed-cell foams¹⁰, which had not been reported to the United Nations Environment Programme (UNEP)^{3,4}.

The previously reported⁴ increase in emissions of 7 ± 3 Gg yr⁻¹, for the period 2014-2017 compared with 2008-2012, from a region denoted “eastern mainland China”, has been revised slightly to 7 ± 4 Gg yr⁻¹ in this study (see Methods). The estimate was made by analysis of observations from the AGAGE Gosan station on Jeju Island, South Korea, and the National Institute for Environmental Studies (NIES) station on Hateruma Island, Japan. The emissions increase was centred on

Shandong and Hebei provinces; Anhui, Beijing, Jiangsu, Liaoning, Shanghai, Tianjin and Zhejiang provinces were included in the eastern mainland China region, but showed lower or near-zero increases, or decreases, in emissions. An increase in emissions from eastern China has since been confirmed by independent atmospheric data from Taiwan¹². In addition, Chinese authorities have reported that, during inspections of several industrial facilities, small quantities (tens of tonnes) of ozone-depleting substances (ODSs), including CFC-11, had been seized, suggesting new production of CFC-11^{5,6}.

In 2018 and 2019, data from Gosan and Hateruma indicate a decline in emissions from eastern Asia (Figure 1). This conclusion can be drawn from the decrease of atmospheric CFC-11 mole fraction enhancements above background levels after 2017, suggesting a fall in emissions from regions to which the monitoring stations are sensitive (see Extended Figure 1). Of the two stations, Gosan is more strongly influenced by industrialised sources in north-eastern China, where the post-2012 emission rise was centred. Unfortunately, substantial periods of instrumental down-time occurred at Gosan in 2016, 2017 and 2018 as a result of typhoon-related damage. These periods of missing data, combined with seasonal changes in meteorology, make it difficult to determine whether emissions changed suddenly, or more gradually throughout 2018 and 2019 (see Methods).

The quantification and attribution of emissions on regional scales using atmospheric mole fraction data by inverse modelling requires models of atmospheric transport that account for meteorological influences on the observed CFC-11 mole fractions. Here, we update inverse estimates of CFC-11 emissions from eastern Asia using Gosan and Hateruma data, based on two atmospheric chemical transport models, NAME and FLEXPART (see Methods), and four independent statistical approaches (denoted NAME-HB, NAME-InTEM, FLEXPART-MIT and FLEXPART-Empa)⁴. Consistent with the observed decline in above-baseline mole fractions (Extended Figure 2), all modelling approaches derive a drop in CFC-11 emissions after 2017 (Figure 2); multi-inversion mean emissions in 2019 were 10 ± 3 Gg yr⁻¹ lower than 2014 - 2017. Emissions during 2018 appear to be intermediate between those derived for 2014-2017 and 2019, which may suggest a gradual reduction in emissions between 2018 and 2019, or could imply a step-change at some point during 2018 (see Methods). Average emissions for the year 2019 from the four inverse methods, were 5.0 ± 1.0 Gg yr⁻¹, which is consistent, within 1-sigma uncertainty,

with the emissions of 7.2 ± 1.5 Gg yr⁻¹ inferred for the period 2008 - 2012 (i.e. before the new emissions rise). Similar to our previous study⁴, these values represent a multi-inversion average, assuming fully correlated uncertainties (see Methods). Analysis of the spatial distribution of the derived emissions reveals that the maximum over Shandong and Hebei between 2014 - 2017 is largely absent in 2019 (Figure 3 and Extended Data Figure 3).

The revised 7 ± 4 Gg yr⁻¹ estimated increase in emissions in 2014-2017 compared with 2008-2012 from eastern China explains $60 \pm 40\%$ of the global emissions rise over the same period (the global increase has also been revised using a correction for the influence of atmospheric dynamics¹³). As in eastern Asia, global background CFC-11 atmospheric monitoring data provide evidence that, in 2019, global emissions returned to levels consistent with those observed before 2013¹³. Our estimated emissions reduction of 10 ± 3 Gg yr⁻¹ from eastern China between 2014-2017 and 2019 accounts for $60 \pm 30\%$ of the global decline during the same period. We do not know where the remaining emissions changes have occurred, as demonstrated in our previous study, which found no evidence of an emission increase in western North America, north-west Europe or southern Australia, where AGAGE stations are located⁴. This result highlights that substantial changes in the emissions of key ODSs have likely occurred in regions not directly sensed by downwind atmospheric measurement stations.

Assuming that unreported CFC-11 production in eastern mainland China started in 2013 and ended in 2018, we can make an estimate of the excess emissions above those originating only from banks (see Methods). Based on our inferred emissions estimates, integrated excess emissions between 2013 and 2018 (inclusive) were 46 (34-60) Gg (median and 95% uncertainty) from eastern China, or 42 (28-56) Gg with a more constrained production scenario (see Methods). This estimate includes fugitive emissions during production of CFC-11, emissions that occurred at the point of use (e.g. as a blowing agent), and those resulting from any addition to, and subsequent emission from the bank (see Methods).

Two substances whose emissions were historically related to CFC-11 production¹⁰ are CCl₄, the chemical feedstock for CFC-11, and CFC-12, which is typically co-produced with CFC-11. Emissions of CCl₄ had increased from north-eastern China around the same period as the rise in

CFC-11 emissions¹⁴. However, its emissions are known to be associated with several industrial activities, such as the manufacture of other chloromethanes, so it is not possible to conclusively attribute this change to CFC-11 production^{15,16}. The ratio of co-produced CFC-12 to CFC-11 can be highly variable¹⁰ as the emissions ratio of CFC-12 relative to CFC-11 depends on whether the CFC-12 was vented to the atmosphere, destroyed, stockpiled, or banked in new equipment. Previous investigation did not find evidence for an abrupt increase in CFC-12 emissions since 2013, although a detailed inverse analysis was not carried out⁴. Notwithstanding the above caveats, further information about CFC-11 production may be gleaned through analysis of CFC-12 and CCl₄ emissions from eastern China, even though the timing and location of the recent increase and decrease in regional CFC-11 emissions need not necessarily coincide with those of its production.

Inverse modelling estimates of CCl₄ and CFC-12 emissions from eastern China using Gosan data show similarities to the changes in CFC-11 emissions between 2008 and 2019 (Figure 2 and Extended Data Figure 4; Hateruma observations were not available throughout this period for these compounds). All three compounds exhibit a small peak in emissions in 2010, perhaps related to the onset of Montreal Protocol restrictions, although, due to the lack of emissions data before 2008, we cannot infer whether pre-2010 emissions are anomalously low or 2010 emissions are larger than expected. In contrast to bottom-up estimates, inverse modelling-derived emissions of CCl₄ increased after 2012, growing from 6.0 ± 1.4 Gg yr⁻¹ for 2011 - 2012 to 10.9 ± 2.0 Gg yr⁻¹ for 2014 - 2017. These emissions subsequently declined around 2017, returning to 6.2 ± 1.4 Gg yr⁻¹ in 2018 - 2019, similar to the 2011 - 2012 estimates. Consistent with our earlier qualitative findings⁴, eastern China emissions of CFC-12 did not change significantly between 2011 - 2012 and 2014 - 2017 (3.0 ± 1.2 Gg yr⁻¹, compared to 3.1 ± 1.2 Gg yr⁻¹, respectively). However, CFC-12 emissions did decline substantially after 2016, reaching 0.7 ± 0.9 Gg yr⁻¹, indistinguishable from zero, for 2017 - 2019.

One possible explanation for the CFC-12 emissions trend is that the expected decline in emissions from CFC-12 banks¹⁷ was approximately equal to the increasing production-related emissions, which then ceased around 2017, and thus no appreciable emissions change was observed before 2017. Under this assumption, we find cumulative excess emissions of 8 ± 4 Gg for CFC-12 between 2013 and 2018 (inclusive, see Methods). For CCl₄, overall enhanced emissions were 23

± 6 Gg during this period. Our results show that emissions of CCl₄ and CFC-12 dropped either one (CCl₄) or two (CFC-12) years before those of CFC-11, which may be consistent with a fall in CFC-11 production before stockpiled CFC-11 was used. The possibility of an increase in CFC-11 emissions was reported at scientific meetings from May 2016 onwards^{18,19}, two years before the first peer-reviewed publication³. Therefore, it is possible that a drop in production during 2017 (as implied by CFC-12 emissions trends), or 2018 (as implied by CCl₄) could have followed from the first public dissemination of anomalous background CFC-11 trends.

A simple process model and some simplifying assumptions can be used to determine the level of CFC-11 production in eastern China that is consistent with our estimates of integrated excess emissions of CFC-11, CFC-12 and CCl₄ during 2013-2018 (see Methods). A recent assessment¹⁰ found that CFC-11 was most likely used to manufacture closed-cell foams, with a release fraction of 25-35% of the produced CFC-11. Using these findings to constrain the process model, unreported production may be up to 165 Gg (median of 118 Gg, lower bound of 74 Gg), with an increase in the 2019 bank of up to 112 Gg (median of 75 Gg, lower bound of 46 Gg; Extended Data Table 2). These results are also supported by an estimate with no explicit assumptions about the target species during production nor its usage application (Extended Data Table 3), which suggest that total unreported production of CFC-11 was up to 109 Gg between 2013-2018 (95% uncertainty interval upper bound, with a median of 62 Gg and lower bound of 44 Gg). This finding results in an increase in the bank size in 2019 of up to 58 Gg (lower bound of 0 Gg, median of 14 Gg), reflecting a short-term CFC-11 release during its production and at the point of use (e.g. during foam manufacture) of between 40% and 100% of the total amount produced.

The integrated excess emissions due to new production, use and increased bank size between 2013 and 2018 for all three species are equivalent to 71 (52-90) Gg of CFC-11 emissions (median and 95% uncertainty, see Methods) in terms of the impact on stratospheric ozone and 362 (247-479) Tg CO₂ in terms of the impact on climate over a 100-year time horizon, equivalent to around 4% of China's 2018 fossil fuel CO₂ emissions²⁰. If we assume that all of the inferred CFC-11 production within eastern China will ultimately be released to the atmosphere, and include the associated excess emissions of CCl₄ and CFC-12 already seen, total Antarctic ozone loss associated with these additional ODS emissions⁸ will be less than 0.2% (see Methods). Therefore,

we can conclude that substantial future stratospheric ozone depletion from renewed, unreported production of CFC-11 and associated ODS from eastern China has been avoided, perhaps instigated by the timely reporting of increased emissions. Coincident with our findings for eastern China, there is evidence that a return to an emissions trajectory more consistent with Montreal Protocol regulations has also taken place globally¹³. These findings underscore the rapid progress that has been made to address this important issue at both regional and global scales. However, due to gaps in the global ODS monitoring system, it remains unclear which other parts of the world contributed to $40 \pm 40\%$ of the global emission rise, or $40 \pm 30\%$ of the subsequent fall, that could not be attributed to eastern China.

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274 **Figure 1: Observations of atmospheric CFC-11 at Gosan and Hateruma.** Dry-air mole fractions
 275 measured for CFC-11 in discrete air samples at Gosan, Jeju Island, South Korea (33.3° N, 126.2° E) and
 276 Hateruma, Japan (24.1° N, 123.8° E) used in the study, from 1 January 2008 to 31 December 2019. The
 277 grey lines show the mole fraction observations at Cape Grim, Tasmania, Australia (40.7° S, 144.7° E) for
 278 comparison.

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Figure 2: Estimated annual mean emissions of CFC-11, CFC-12 and CCl₄ for eastern mainland China. Annual mean emissions are shown for (a) CFC-11, (b) CFC-12 and (c) CCl₄, where the solid black line shows the mean estimate of the four inversion frameworks and the shading shows the estimates for each inversion within their one standard deviation or 68% uncertainty. Inventory-based estimates of emissions for all four gases from eastern mainland China (determined as the total Chinese emissions scaled by the fraction of the population, 35%, that reside in that part of the domain) are shown as a dashed line (including projected inventory values¹⁷ after 2014). For CCl₄, additional bottom-up estimates are shown as a dotted line with black squares with the associated 95% uncertainty²¹, and by the black diamond¹⁶, also scaled by population.

Figure 3: **Mean spatial distribution of the mean CFC-11 fluxes from the four inversions.** Emissions are shown for (a) 2008-2012, (b) 2014-2017, and (c) 2019. The black triangle and circle indicate the Gosan and Hateruma stations, respectively. The hatched areas indicate regions to which the observations have low sensitivity and are therefore excluded from the emissions presented in the main text⁴. Derived emissions distributions are similar for each inversion (Extended Data Figure 3).

Methods

Atmospheric observations of CFC-11, CFC-12 and CCl₄

CFC-11 observations were updated through 2019 using the same methods presented previously from Gosan, South Korea and Hateruma, Japan⁴. Mole fraction measurements of CFC-12 and CCl₄ were only available from Gosan. All measurements were made using gas-chromatography mass-spectrometry (GC-MS)^{22,23}, with ~12 measurements per day at Gosan, and ~20 measurements per day at Hateruma. Measurements at Gosan are on the Scripps Institution of Oceanography (SIO) SIO-05 calibration scale, at Hateruma on the NIES-08 calibration scale. Hateruma CFC-11 data were converted to the SIO-05 scale by multiplying by an intercalibration factor of 0.996, derived through inter-laboratory air sample exchange.

Gosan measurement station has several large data gaps in 2016, 2017 and 2018 due to periods of instrumental down-time as a result of typhoon-related damage. The prevailing winds at both measurement stations, and therefore when air from a particular region is sampled, are seasonally dependent. Compounded by missing data, this makes it difficult to determine when sub-annual emissions changes occurred.

Inverse modelling framework

Our inverse estimates use atmospheric chemical transport models and Bayesian methods to infer source magnitudes from atmospheric measurement data. We used two backward-running Lagrangian models, the UK Met Office Numerical Atmospheric-dispersion Modelling Environment (NAME)²⁴ and the FLEXible PARTicle dispersion model (FLEXPART)^{25,26} to estimate the sensitivity of the observations to trace gas emissions. These models were used with four inversion frameworks developed and run independently by four groups: the University of Bristol's hierarchical Bayesian inversion (NAME-HB)^{27,28} and the Met Office's Inversion Technique for Emissions Modelling (NAME-InTEM)^{29,30}, which used NAME, and an analytical inversion framework developed by MIT (FLEXPART-MIT)^{31,32} and Empa's Bayesian regional inversion system (FLEXPART-Empa)^{33,34}, which used FLEXPART. The model set-up used in each inversion framework, including its meteorological inputs, are detailed in Extended Data Table 1.

For CFC-11, previous work details how these methods have been applied⁴. They are briefly outlined below, with some minor changes, which have led to a small revision of the previously published emissions rise from eastern China after 2013 (main text). The inversions for CFC-12 and CCl₄ were run with the same statistical methodology as for CFC-11, and the same model derived sensitivities of the atmospheric measurements to emissions. Emissions were inferred for the same eastern mainland China region for CFC-12, CCl₄ and CFC-11. Previous work⁴ has shown that inferred emissions from this region using

measurement data from only Gosan are consistent with emissions inferred using measurements from both Gosan and Hateruma, albeit with larger uncertainty (comparison updated here in Extended Data Figure 5). All inversion results presented here are based on *a priori* emissions estimates distributed uniformly in space within each country, or within particular domains (see below), which did not change from year to year. Therefore, the derived emissions trends were informed only by the data. As in the previous study⁴, emissions were also estimated using *a priori* emissions distributed by population to show that the results are insensitive to the choice of the emissions *a priori* spatial distribution (Extended Data Figure 6). The residual between the models and the data was similar for each inversion approach, and did not change appreciably throughout the study period (Extended Data Figure 7).

The NAME-HB inversion for CFC-11 has been outlined previously⁴. Here we show updated results for 2018 and 2019. The inversion is not performed on a fixed underlying grid, but a variable basis function representation, which is explored during the inversion²⁸, where the highest allowable spatial resolution is the NAME output resolution. The treatment of the measurement data averaging, model-measurement error and prior constraints on the scaling of the *a priori* emissions and boundary conditions were the same for all three gases. *A priori* estimates of CFC-12 for all years were 20 Gg yr⁻¹ in the inner domain, which were distributed uniformly over land (not water), and 20 Gg yr⁻¹ in the outer domain (see our previous study⁴ for definitions of these domains). For CFC-12, *a priori* mole fraction concentrations at the domain boundary were equal to the monthly baseline mole fraction derived from measurements at the Mace Head measurement site in Ireland. The NAME-HB *a priori* emissions estimates for CCl₄ for each country were taken from an inventory estimate for 2014¹⁶ where available (Russia 0.6 Gg yr⁻¹, China 7.3 Gg yr⁻¹, India 2.8 Gg yr⁻¹, Japan 0.6 Gg yr⁻¹, South Korea 0.2 Gg yr⁻¹), or else they were taken to be 0.05 Gg yr⁻¹, with the emissions distributed uniformly over land within each country. The *a priori* mole fractions at the boundary of the domain uses climatological estimates from the NASA 3D GEOS Chemistry Climate model (as for CFC-11)³⁵.

The NAME-InTEM inversions for CFC-11, CFC-12 and CCl₄ used uniform *a priori* estimates over land within the inversion domain (16.0°N - 57.6° N, 88.1° E - 145.9° E) of 50 Gg yr⁻¹ (CFC-11, CFC-12) and 30 Gg yr⁻¹ (CCl₄). These were given large uncertainties resulting in estimates of total emissions of 23 (0–147) Gg yr⁻¹ for China, 2.6 (0–26) Gg yr⁻¹ for eastern mainland China, and 1.2 (0–8.6), 0.3 (0–3.4) and 0.4 (0–3.9) Gg yr⁻¹ for western Japan (using a previous definition of western Japan⁴), South Korea and North Korea, respectively, for CFC-11 and CFC-12. For CCl₄, the *a priori* estimates were 14 (0–90) Gg yr⁻¹ for China, 1.6 (0–16) Gg yr⁻¹ for eastern mainland China, and 0.7 (0–5), 0.2 (0–2) and 0.2 (0–2.3) Gg yr⁻¹ respectively for western Japan, South Korea and North Korea. *A priori* emissions over water were set

to zero and assigned a smaller prior uncertainty relative to emissions on land. The methodology of the NAME-InTEM model has been previously reported⁴, however there have been adjustments to some of the modelling parameters which have resulted in changes to the estimated NAME-InTEM CFC-11 compared to our previous study. The observations were averaged into 6 h inversion windows and the observational uncertainty was time varying and estimated as the variability of the observations in the 12 h period centred on the inversion window plus the measurement repeatability determined from repeat measurements of the on-site air tank. Model uncertainty was estimated every 6 h as the larger of the median of all pollution events at each station in a year or 10% of the magnitude of the pollution event. A temporal correlation of 12 h was assumed in the model uncertainty at each station. An analytical solution was found that minimized the residual between the model and the observations and the difference between the posterior and prior flux estimates, balanced by the uncertainties of both. The mole fraction baseline at each station was estimated as described previously^{4,30}. The inversion grid has a variable resolution, which was calculated within InTEM based on the magnitude of the footprint and the emissions from each region⁴. If there is sufficient signal from a region, the model divides the region and recalculates, down to a minimum grid size of 25 km by 25 km.

The FLEXPART-MIT inversion derived fluxes for CFC-11, CFC-12 and CCl₄ used FLEXPART with an analytical Bayesian framework³². *A priori* flux fields were set to be spatially uniform over continental eastern Asia, and not allowed over water. Emissions were solved within fixed variable-resolution grid cells with the finest grid (1° x 1°) in eastern China and other eastern Asian countries, with a coarsest grid resolution of 24° x 24°. *A priori* estimates of CFC-11 fluxes were 14.5 Gg yr⁻¹ for China (1.5 Gg yr⁻¹ for eastern mainland China) and 1.2, 0.27 and 0.27 Gg yr⁻¹ for Japan, South Korea and North Korea, respectively. For CFC-12 and CCl₄, the *a priori* flux estimates were 14.5 and 7.7 Gg yr⁻¹, respectively. The uncertainty on each grid cell was arbitrarily set to 1000% of the *a priori* estimate and the spatial correlation length was assumed to be 300 km. Baseline mole fractions were estimated at each site every 7 days. Model-measurement uncertainty for each 24-hour-averaged observation was estimated using the quadratic sum of: 1% of the baseline value (as a measure of baseline uncertainty); the measurement repeatability; and the standard deviation of the 24-hour variability (as a measure of the model-data ‘mismatch’ uncertainty). Similar to InTEM, *a posteriori* fluxes were derived using an analytical approach that minimizes a cost function that weights the measurement-model residuals and the *a priori* estimate by their respective uncertainties.

FLEXPART-Empa inversions for CFC-11 used the same model setup as previously described⁴. The inversion system uses a fully Bayesian approach, pre-calculating hyper-parameters that describe the

covariance structure by applying maximum likelihood optimisation³³. Emissions were estimated on a rectangular grid with variable resolution for which the grid spacing was approximately inversely proportional to average source sensitivities. The resulting grid cell sizes ranged from 0.125° x 0.125° to 8° x 8°. The inversion domain comprised the area from 80°E to 140°E and 20°N to 60°N. *A priori* emissions for CFC-12 were set homogeneously over land areas in the inversion domain of eastern Asia, totalling 29 Gg yr⁻¹ for all years. The model setup for CCl₄ followed that for CFC-11 and CFC-12 with the difference of *a priori* emissions being taken from an inventory estimate for 2014¹⁶ where available (China 7.3 Gg yr⁻¹, Japan 0.6 Gg yr⁻¹, South Korea 0.2 Gg yr⁻¹) and set to 0.05 Gg yr⁻¹ per country elsewhere in the inversion domain. Again, the emissions were distributed uniformly over land within each country. Emissions were allowed over ocean-only grid cells, but were assigned smaller *a priori* emissions (factor 100 smaller than over land) and smaller prior uncertainty relative to land cells.

Multi-inversion means were estimated as previously⁴. We assume that the emissions estimates from individual models for each year are fully correlated (not independent) due the fact that the underlying physics used to derive the sensitivities of the measurements to emissions in FLEXPART and NAME is similar, as are the observations and physics used to derive their meteorological drivers. We do not explore these systematic uncertainties in the inversions, and so they are not reflected in the posterior uncertainty estimates.

Estimation of CFC-11 production and excess emissions

We estimated the magnitude of CFC-11 production between 2013 and 2018, and the “excess” emissions above those expected due to release from the pre-2013 bank alone. To do this, we assume a simplified model of CFC-11 production, with CCl₄ as a starting reagent and CFC-12 as a byproduct. We assume that some fraction of the CCl₄ is released to the atmosphere during its production and during the production of CFC-11, that the bank release fraction for CFC-11 does not change with time and that the magnitude of the addition to the bank is constant during each year between 2013 and 2018.

In this framework, the annual CFC-11 emissions in 2012 (Q_{CFC-11}^{2012} in moles) are only from the banks (B_0),

$$Q_{CFC-11}^{2012} = B_0 f_b \quad (1),$$

where f_b is the bank release fraction. We assume that renewed production ceased in 2018. Therefore, the CFC-11 emissions in 2019 are a combination of remaining emissions from the pre-2013 bank and the bank accumulated during the N years of renewed production, i.e. the 6 year period 2013-2018,

$$Q_{CFC-11}^{2019} = B_0 f_b (1 - f_b)^{N+1} + (1 - f_{CFC-11}) \frac{P_{CFC-11}}{N} f_b \sum_{i=1}^N (1 - f_b)^i, \quad (2)$$

where f_{CFC-11} is the fraction of CFC-11 released promptly to the atmosphere at the time of use or during production and P_{CFC-11} is the cumulative production of CFC-11 between 2013 and 2018 in moles. The integrated total emissions between 2013-2018 are a combination of emissions from the pre-2013 bank, the additional bank from renewed production and emissions from prompt release during CFC-11 production and usage,

$$Q_{CFC-11}^{2013-2018} = B_0 f_b (1 - f_b)^N + (1 - f_{CFC-11}) \frac{P_{CFC-11}}{N} f_b \sum_{i=0}^{N-1} (1 - f_b)^i + P_{CFC-11} f_{CFC-11}. \quad (3)$$

The excess emissions of CFC-12 (see next section for description of how this was calculated), assuming that no recently produced CFC-12 was added to its bank, is related to the amount of CFC-11 produced through

$$Q_{CFC-12}^{excess} = P_{CFC-11} f_{CFC-11/CFC-12}, \quad (4)$$

where the term $f_{CFC-12/CFC-11}$ accounts for the fraction of CFC-12 that was co-produced with CFC-11 and any CFC-12 that was banked or destroyed after production.

Excess emissions of CCl_4 are related to the total amount of CFC-11 and CFC-12 produced by

$$Q_{CCl_4}^{excess} = P_{CFC-11} (1 + f_{CFC-11/CFC-12}) f_{CCl_4}, \quad (5)$$

where f_{CCl_4} is the overall fraction of CCl_4 that is emitted to the atmosphere during its production, and during its transport and use as a CFC-11 feedstock.

This system of equations is underdetermined and therefore additional constraints are needed. We investigated two solutions, one that only constrains the uncertain parameters relating to CFC-11 production to be within physical bounds, while the other uses likely ranges from a recent assessment¹⁰. In the first case, we made no assumptions about whether the target species of production is CFC-11 or CFC-12, nor about its usage (e.g. closed or open cell foams), and so provided uninformative constraints on the unknowns $f_{CFC-12/CFC-11}$, f_{CCl_4} and f_{CFC-11} , allowing them to be between 0 and 100% (with a uniform prior distribution). In the second case, we included an assessment of likely production and usage¹⁰, which constrained $f_{CFC-12/CFC-11}$ to 0 - 30%, assuming that CFC-11 is the target production species, and f_{CFC-11} to 25 - 35%, assuming that it is used in closed-cell foam manufacturing, with a uniform prior distribution. The CFC-11 production (P_{CFC-11}) was allowed to vary from between the integrated 2013-2018 emissions in excess of a linear fit between the 2012 and 2019 emissions, to a factor of 100 times larger (uniform distribution). The results were not sensitive to the precise value of this upper limit, which was found to be far outside the posterior uncertainty range). The prior probability of the pre-2013 bank B_0 was given a uniform distribution with a lower bound of Q_{CFC-11}^{2012} and an upper bound 100 times larger than Q_{CFC-11}^{2012} . The bank release fraction f_b was given a uniform prior distribution with a lower bound of 1.5% and an upper bound of 4.2%¹⁰. By using

the prior constraints on the unknown variables, although only partly informative, a Bayesian probabilistic framework allows full exploration of the possible production ranges, given the inferred emissions, their uncertainties and our prior assumptions. The system of equations was solved using a Hamiltonian Markov Chain Monte Carlo method and the PyMC3 library^{36,37}.

The derived parameter values using emissions data estimated by each inversion, and using the mean emissions from all inversions, are shown in Extended Table 3, using no assumptions about the target production species nor its application, and Extended Table 2 under the most likely scenario for production and usage. The values derived from the multi-inversion mean are consistent with those of the individual inversions. Therefore, we use the multi-inversion mean in the main text. Production values for CFC-11 and its prompt release fraction are discussed in the main text. We infer a co-production of CFC-12 of 0 - 30% (95% uncertainty range), which is in line with expectations, if CFC-11 was the main target of new production¹⁰. A relatively high overall fugitive release rate of CCl₄ of 8 - 49% (due to CCl₄ production, transport and CFC-11 production) is implied. The higher end of this range seems unlikely and could be indicative of a coincident change of some other process relating to CCl₄ emissions³⁸, potentially including the production of chloroform (CHCl₃), which increased between 2010 and 2015³². Using the likely production and usage scenario, co-production of CFC-12 is constrained to 0 - 15%, with a fugitive CCl₄ release rate of 5.8 - 26.9%.

We define “integrated excess” CFC-11 emissions as the cumulative total emissions above those that originated only from the inferred pre-2013 bank. Therefore, our integrated excess emissions include any CFC-11 that was released promptly during production and usage and any that was added to the bank and subsequently emitted between 2013 and 2018.

Calculation of CCl₄ and CFC-12 excess emissions

It was assumed that, without the new CFC-11 production, the emissions of CFC-12 and CCl₄ would have followed a linear trend between 2012 and 2019 and the 2019 emissions were not influenced by any additional contribution to the CFC-12 or CCl₄ bank. Integrated excess emissions can therefore be approximated as the sum of annual emissions above a trend line fitted between the 2012 and 2019 emissions. A Monte Carlo uncertainty estimate used an ensemble of emissions estimates, where a normally distributed random number was added to the emissions in each year, with a standard deviation equal to the multi-inversion uncertainty. For each ensemble member, the trend and excess emissions were calculated. Values presented in the main text are the mean and standard deviation of 100,000 ensemble members. Emissions for CFC-12 and CCl₄ relied on data from Gosan only, whereas CFC-11 estimates were inferred from Gosan

and Hateruma data together. Therefore, it is possible that integrated excess emission estimates could be somewhat different for CCl₄ and CFC-12, if Hateruma data were available, although whether they would be higher or lower will likely be dependent on the relative spatial distributions of the emissions for these species.

Calculation of impact on stratospheric ozone and climate

The global warming potentials on 100-year time horizon (GWP₁₀₀) of CFC-11, CFC-12 and CCl₄ were 5160, 10300 and 2100 respectively³⁹, which equates to 362 (247-479) Tg CO₂ using no assumptions about production and usage, or 343 (231-457) Tg CO₂ for the most likely case. The ozone depletion potentials were 1, 0.77 and 0.89 for CFC-11, CFC-12 and CCl₄ respectively, where the ozone depletion potential for CFC-12 was assumed to be the mean of the range presented in the literature³⁹. Using a linear relationship between CFC-11 emissions and stratospheric ozone loss⁸, the total impact on stratospheric ozone was estimated by converting the derived excess emissions for all gases into a CFC-11-equivalent total. The cumulative emissions from eastern China alone – 71 (52-90) Gg CFC-11-equivalent in terms of the impact on stratospheric ozone using no production nor usage assumptions – would account for a reduction over the coming decades in global column ozone, or spring-time Antarctic ozone, of only around 0.007% or 0.07%, respectively. The same ozone reduction would be observed using the most likely production case. If we assume that all of the inferred CFC-11 production within eastern China will ultimately be released to the atmosphere, and including the temporary excess emissions of CCl₄ and CFC-12, total associated ozone loss under no production and usage assumptions (or the most likely production case) would be between 0.006 - 0.01% (0.01 - 0.02%) globally or 0.06 - 0.1% (0.1 - 0.2%) for the Antarctic spring.

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Author contributions

S. P., T. S., P. F., C. H., J. K., P. K., J. M., S. O'D., M.-K. P., P. S. and R. F. W. provided observations and data quality assurance. L. W., A. R., S. H., X. F., A. J. M., R. G. P., A. L. G., Q. L., and M. L. R. provided model output or carried out inverse modelling. All authors wrote the paper.

Competing interests

The authors declare no competing interests.

Data availability

Data from Gosan and Cape Grim stations are available from the AGAGE website (<https://agage.mit.edu>). Hateruma data are available from the World Data Centre for Greenhouse Gases (<https://gaw.kishou.go.jp/>). Data pertaining to the emissions of CFC-11, CFC-12, and CCl₄ are available from the OSF repository 'Data and code for emissions of CFC-11 and related substances from eastern China' (<https://osf.io/qp2be/>, DOI 10.17605/OSF.IO/QP2BE).

Code availability

Licences to use NAME and InTEM are available for research purposes via a request to the UK Met Office or on request from A.J.M. and A.L.R. The code for the NAME-based hierarchical Bayesian inversion (NAME-HB) is available on request from M.R. and L.M.W. The code of the dispersion model FLEXPART is available from www.flex-part.eu. The code for the FLEXPART-based Bayesian inversion (FLEXPART-

MIT) is available on request from X.F. The inversion code used by Empa is available from <https://doi.org/10.5281/zenodo.1194642> or on request from S.H. The algorithm used to estimate the excess emissions of CFC-11, CFC-12 and CCl₄, and the production of CFC-11, for eastern China is available from the OSF repository ‘Data and code for emissions of CFC-11 and related substances from eastern China’ (<https://osf.io/qp2be/>, DOI 10.17605/OSF.IO/QP2BE).

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623 **Extended data**

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625 **Extended Figure 1: The mean sensitivity of the measurements to emissions of CFC-11.** Each panel
626 shows the sensitivity of the measurements to emissions of CFC-11 for each year between 2008-2019. The
627 black triangle and circle indicate the Gosan and Hateruma stations, respectively. The sensitivities are
628 derived using the NAME model for daily averaged measurement values at each station. Where no
629 measurement data were available from a station, this is not included in the mean sensitivity. The figure
630 shows that the mean sensitivity of the observations to emissions from eastern China did not change
631 substantially throughout this period.

Extended Figure 2: Annual mean above-baseline enhancement in the mole-fraction measured at Gosan for trajectories originating from China. Annual mean above-baseline mole-fraction enhancements are compared to emissions estimates from eastern mainland China from the four inversion systems for (a) CFC-11, (b) CFC-12 and (c) CCl₄. The shading shows the estimated emissions for each inversion within their one standard deviation or 68% uncertainty and the black line shows the mean enhancement in mole fraction from China measured at Gosan, with the one standard error variability shown by the black dotted line. Baseline mole fractions were determined using a statistical method⁴⁰, and air masses were classified as originating from China where above-baseline pollution events arriving at Gosan had entered the boundary layer only within the Chinese country domain within their 6-day kinematic back trajectories. Back trajectories were calculated using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model⁴¹ of the NOAA Air Resources Laboratory (ARL) using meteorological information from the Global Data Assimilation System (GDAS) model with 1°×1° grid cell.

Extended Figure 3: **Maps of the mean CFC-11 fluxes from the four inversions.** Spatial distribution of the mean CFC-11 fluxes from the four inversions: (a)-(c) NAME-HB, (d)-(f) NAME-InTEM, (g)-(i) FLEXPART-MIT, and (j)-(l) FLEXPART-Empa. The plots show the average emissions from 2008-2012 in (a), (d), (g) and (j); the average emissions for 2014-2017 in (b), (e), (h) and (k); and the emissions for 2019 in (c), (f), (i) and (l). The black triangle and circle indicate the Gosan and Hateruma stations, respectively. The hatched areas indicate regions of the domain to which the observations have low sensitivity, and therefore, from which the derived emissions have high uncertainty⁴. As a result, only emission magnitudes and emission changes for the non-hatched regions are included in the values quoted in the main text.

Extended Figure 4: **Maps of the mean flux averaged over the four inversions for CFC-11, CFC-12 and CCl₄** The mean spatial distribution of the mean fluxes are for (a), (b) and (c) CFC-11; (d), (e) and (f) CFC-12; and (g), (h) and (i) CCl₄ from the four inversions for the periods: (a), (d) and (g) 2008-2012; (b), (e) and (h) 2014-2017, and (c), (f) and (i) 2019. The black triangle and circle indicate the Gosan and Hateruma stations, respectively, which are the measurement sites used to derive the emissions. The hatched areas indicate regions of the domain to which the observations have low sensitivity, and therefore, from which the derived emissions have high uncertainty⁴. Only emission magnitudes for the non-hatched regions are included in the values quoted in the main text. The different spatial distributions for the three gases reflect different emissions distributions from their respective banks (primarily for CFC-11 and CFC-12), or differences in production-related emissions (e.g. the production of chloromethanes is thought to be a substantial source for CCl₄, but not for CFC-11 and CFC-12¹⁶).

Extended Data Figure 5: **A comparison of emissions estimates for eastern mainland China using different measurement datasets.** Estimated annual emissions of CFC-11 using *a priori* emissions distributed uniformly over land using measurements from (a) Gosan and Hateruma, (b) Gosan and (c) Hateruma stations. The black line shows the mean estimate of the four inversion frameworks and the shading shows the estimates for each inversion within their one standard deviation or 68% uncertainty. Inventory-based estimates of emissions for all four gases from eastern mainland China (determined as the total Chinese emissions scaled by the fraction of the population, 35%, that reside in that part of the domain) are shown as a dashed line (including projected inventory values¹⁷ after 2014).

Extended Data Figure 6: **Emissions estimates for eastern mainland China using *a priori* emissions distributed by population.** Estimated annual emissions of (a) CFC-11, (b) CFC-12 and (c) CCl₄ for eastern mainland China using *a priori* emissions distributed by population in space. The black line shows the mean estimate of the four inversion frameworks and the shading shows the estimates for each inversion within their one standard deviation or 68% uncertainty. Inventory-based estimates of emissions for all four gases from eastern mainland China (determined as the total Chinese emissions scaled by the fraction of the population, 35%, that reside in that part of the domain) are shown as a dashed line (including projected inventory values¹⁷ after 2014). For CCl₄, the black symbols show additional bottom-up estimates, shown by a dotted line and black squares with the associated 95% uncertainty²¹; and by the black diamond¹⁶, also scaled by population. The derived emissions are consistent with those derived in the main text, which assume *a priori* emissions that are spatially uniform over land.

Extended Figure 7: **Simulated and observed CFC-11 mole fractions for the four inverse frameworks.** Left, a comparison of the simulated CFC-11 mole fractions from the four different inversion analyses and those that were measured at Gosan and Hateruma. Right, residuals between the simulated and observed mole fractions (data minus model). Shading denotes the 1-sigma model-data mismatch uncertainties assumed in the inversions. Simulated mole fractions are derived from *a posteriori* emissions. For the NAME-InTEM, FLEXPART-EMPA, NAME-HB and FLEXPART-MIT inversions, 6-hourly, 3-hourly, 24-hourly and 24-hourly averaging was applied to the model and data, respectively, which represents the temporal resolution at which the model simulates the observed data using derived annual emissions.

Method	Lagrangian Particle dispersion model	Number of particles and air history	Model meteorology	Resolution of meteorology	Model domain
NAME-HB	NAME ²⁴	20,000 per 1 h for 30 days	UK Met Office Unified Model ⁴²	3-hourly; from 0.563° x 0.375° to 0.141° x 0.094° (increasing over the study period)	5° S - 74° N and 55° E - 192° E
NAME-InTEM	NAME ²⁴	20,000 per 1 h for 30 days	UK Met Office Unified Model ⁴²	3-hourly; from 0.563° x 0.375° to 0.141° x 0.094° (increasing over the study period)	5° S - 74° N and 55° E - 192° E
FLEXPART-MIT	FLEXPART ^{25,26}	40,000 per 3 h for 20 days	Operational ECMWF analysis ^{43,44}	3-hourly; 1° x 1°	Global
FLEXPART-Empa	FLEXPART ^{25,26}	50,000 per 3 h for 10 days	Operational ECMWF analysis/forecast ^{43,44}	3-hourly; 1° x 1° global resolution and 0.2° x 0.2° resolution for north-eastern China (105° E to 125° E and 30° N to 50° N)	Global

Extended Data Table 1: The atmospheric dispersion model set-up for the four inverse methods, from which the sensitivity ‘footprints’ are derived.

Inversion framework	Excess CFC-11 emissions between 2013-2018 (Gg)	CFC-11 Production (Gg)	Excess added to 2019 CFC-11 banks (Gg)	CFC-11 release fraction from production and use (%)	Fraction of CFC-12 to CFC-11 production (%)	Fraction of CCl₄ emitted (%)	CFC-11 release fraction from bank (%)
Inversion mean	42 (28-56)	118 (74-165)	75 (46-112)	32 (26-35)	7 (0-15)	15 (6-27)	2.2 (1.5-3.8)
NAME-HB	49 (39-59)	137 (103-177)	87 (62-121)	32 (26-35)	6 (3-9)	10 (4-17)	2.1 (1.5-3.7)
NAME-InTEM	42 (18-67)	119 (45-187)	76 (29-126)	31 (26-35)	9 (0-24)	16 (0-37)	2.4 (1.5-3.9)
FLEXPART-MIT	47 (33-62)	132 (86-183)	84 (51-122)	32 (26-35)	5 (0-12)	13 (5-21)	2.2 (1.5-3.8)
FLEXPART-Empa	29 (24-35)	79 (62-100)	50 (37-67)	33 (28-35)	15 (10-20)	27 (18-37)	2.2 (1.5-3.9)

Extended Data Table 2: The estimated parameters related to production of CFC-11 in eastern mainland China from 2013-2018 using the most likely scenario for production and usage. Integrated excess emissions are calculated as the emissions due to new production and use, and the subsequent release from the addition to the banks between 2013-2018 inclusive (see Methods). The reported estimates are the median posterior values and the 95% uncertainty interval using the emissions derived from each inversion framework, and the mean estimate from all inversion frameworks.

Inversion framework	Excess CFC-11 emissions between 2013-2018 (Gg)	CFC-11 Production (Gg)	Excess added to 2019 CFC-11 banks (Gg)	CFC-11 release fraction from production and use (%)	Fraction of CFC-12 to CFC-11 production (%)	Fraction of CCl₄ emitted (%)	CFC-11 release fraction from bank (%)
Inversion mean	46 (34-60)	60 (44-109)	14 (0-58)	73 (40-100)	13 (0-30)	27 (8-49)	2.6 (1.5-4.0)
NAME-HB	50 (41-61)	76 (49-139)	25 (0-84)	63 (32-97)	10 (4-31)	16 (4-31)	2.4 (1.5-4.0)
NAME-InTEM	52 (30-76)	71 (50-144)	21 (0-86)	68 (30-100)	14 (0-41)	24 (0-53)	2.5 (1.5-4.0)
FLEXPART-MIT	50 (37-65)	63 (46-110)	13 (0-55)	77 (43-100)	10 (0-24)	24 (8-42)	2.7 (1.5-4.0)
FLEXPART-Empa	32 (27-37)	39 (31-61)	8 (0-27)	79 (51-100)	31 (18-44)	47 (27-67)	2.9 (1.6-4.2)

Extended Data Table 3: The estimated parameters related to production of CFC-11 in eastern mainland China from 2013-2018 under no assumption of whether CFC-11 or CFC-12 is the target production species, nor its consumption application. Integrated excess emissions are calculated as the emissions due to new production and use, and the subsequent release from the addition to the banks between 2013-2018 inclusive (see Methods). The reported estimates are the median posterior values and the 95% uncertainty interval using the emissions derived from each inversion framework, and the mean estimate from all inversion frameworks.