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1 **Increase in CFC-11 emissions from eastern China based on atmospheric observations**

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4
5 **The recovery of the stratospheric ozone layer relies on the continued decline in the**
6 **atmospheric concentrations of ozone-depleting gases such as the chlorofluorocarbons¹.**
7 **The atmospheric concentration of trichlorofluoromethane (CFC-11), the second most**
8 **abundant chlorofluorocarbon, has declined substantially since the mid-1990s². A**
9 **recently reported slowdown in the decline of the atmospheric concentration of CFC-11**
10 **after 2012, however, implies that global emissions have increased^{3,4}. A concurrent**
11 **increase in CFC-11 emissions from eastern Asia contributes to the global emission**
12 **increase, but the location and magnitude of this regional source remain uncertain³.**
13 **Here we use high-frequency atmospheric observations from Gosan, Republic of Korea**
14 **and Hateruma, Japan, together with global monitoring data and atmospheric chemical**
15 **transport model simulations to investigate regional CFC-11 emissions from eastern**
16 **Asia. We find that emissions from eastern mainland China are 7.0 ± 3.0 Gg yr⁻¹ higher**
17 **in 2014 – 2017 compared to 2008 – 2012, and the emissions increase arises primarily**
18 **around the north-eastern provinces of Shandong and Hebei. This increase accounts for**
19 **a substantial fraction (about 40 – 60%, or more) of the global CFC-11 emission rise. We**
20 **find no evidence for a significant increase in emissions from any other eastern Asian**
21 **countries or other regions of the world where available data allow for the detection of**
22 **regional emissions. Attribution of any remaining fraction of the global CFC-11 emission**
23 **rise to other regions is limited by the sparsity of long-term measurements of sufficient**
24 **frequency near potentially emissive regions. Multiple considerations suggest that the**
25 **increase in CFC-11 emissions from eastern mainland China is likely the result of new**

**1 production and use, inconsistent with the Montreal Protocol agreement to phase out
2 global chlorofluorocarbon production by 2010.**

3

4 Before its global production phase-out in 2010, mandated by the Montreal Protocol on
5 Substances that Deplete the Ozone Layer, CFC-11 was primarily used as an agent for
6 blowing foams that were incorporated into buildings and consumer products. As production
7 declined, it was expected that leakage from banks of CFC-11 stored in these foams would
8 dominate its emissions to the atmosphere. Emissions were projected to decline as these banks
9 depleted¹. However, atmospheric mole fraction data from the National Oceanic and
10 Atmospheric Administration (NOAA) and the Advanced Global Atmospheric Gases
11 Experiment (AGAGE) show that the rate at which CFC-11 has been declining in the global
12 background atmosphere slowed since approximately 2013, and its interhemispheric mole
13 fraction difference increased^{3,4}. Whilst variations in atmospheric circulation may have
14 contributed to the slow-down, an increase in northern hemispheric emissions is required,
15 starting after 2012, to explain the majority of these observed changes³.

16

17 High-frequency atmospheric CFC-11 mole fraction data from the AGAGE network⁴ and
18 Japanese National Institute for Environmental Studies (NIES)⁵ show signals that are
19 consistent with a continuing decline in CFC-11 emissions from Europe, North America and
20 Australia in recent years, but a rise from eastern Asia since 2012 (Figure 1; Extended Data
21 Figure 1). In these datasets, enhancements above the hemispheric background mole fractions
22 are observed when plumes from nearby sources impinge on the measurement stations. For the
23 AGAGE stations in Europe, the west coast of North America, Australia and more remote
24 locations, the magnitudes of above-baseline events are relatively small and have declined
25 throughout the study period (2008 – 2017, inclusive), indicating that emissions near these

1 sites have dropped. A decline in emissions from North America is consistent with a recent
2 study carried out using independent NOAA data in the USA through 2014⁶. In contrast,
3 observations at Hateruma, Japan (location shown in Extended Data Figure 2) show persistent
4 above-baseline events throughout, which have not declined substantially or may have
5 increased, and data from Gosan, Republic of Korea show a strong increase in the magnitude
6 of these events after 2012 (Figure 1). We focus our analysis on eastern Asia because the
7 existing measurement networks only show an increase in the magnitude of above-baseline
8 events in this region. A qualitative emissions increase from somewhere in this region was
9 also indicated in NOAA data from Hawaii³.

10

11 To quantify emissions from eastern Asian countries, atmospheric observation-based
12 “inverse” or “top-down” estimates were carried out with CFC-11 measurements from Gosan
13 and Hateruma observations using two atmospheric chemical transport models and four
14 statistical methods (see Methods). Previous top-down studies in this region between 2001 and
15 2012 had broadly confirmed inventory-based (“bottom-up”) estimates of declining emissions
16 from China^{7–12}, and found comparatively small emissions from Japan and the Korean
17 Peninsula^{10,13}. Here, we used the NAME¹⁴ and FLEXPART¹⁵ models to simulate the
18 transport of CFC-11 from potential sources to the measurement sites. Emissions were derived
19 from the observed mole fractions using NAME and a hierarchical Bayesian “trans-
20 dimensional” inverse method (denoted NAME-HB)¹⁶ and the Bayesian Inversion Technique
21 for Emissions Modelling (NAME-InTEM)¹⁷. Inversions with FLEXPART used two
22 independent analytical Bayesian frameworks (FLEXPART-MIT¹⁸ and FLEXPART-Empa¹⁹).
23 Analysis using NAME shows that the measurements from the Gosan and Hateruma stations
24 are sensitive to recent emissions from western Japan, the Korean Peninsula, and a region that
25 we define as “eastern mainland China”, which encompasses around 35% of the Chinese

1 population and contains the provinces of Anhui, Beijing, Hebei, Jiangsu, Liaoning,
2 Shandong, Shanghai, Tianjin and Zhejiang (Extended Data Figure 2). Hence, emissions
3 totals are subsequently provided here only for these regions of China.
4
5 Between 2008 and 2012 (inclusive), we inferred mean CFC-11 emissions of 6.4 ± 1.2 Gg yr⁻¹
6 for eastern mainland China (Figure 2, the technique for estimating multi-inversion mean
7 emissions is provided in Methods). The mean derived emission during these years for eastern
8 mainland China is similar to an inventory estimate for the whole of China, when scaled by
9 the fraction of the population that reside in the eastern mainland China region⁷. However,
10 whilst the inventory-based emission estimates for China show a downward trend during this
11 period, emission magnitudes derived from atmospheric observations did not decline
12 significantly. After 2012, the top-down estimates from four different inverse techniques show
13 an emission increase of CFC-11 from eastern mainland China, although the exact timing of
14 the initial rise differs between inversions. Average emissions between 2014 and 2017 were
15 13.4 ± 1.7 Gg yr⁻¹ (for consistency with a previous study, we have omitted 2013 as a
16 transitional year³). Therefore, the increase in derived emissions between the periods 2008 –
17 2012 and 2014 – 2017 is 7.0 ± 3.0 Gg yr⁻¹, or around 110 %. Our top-down inversion-based
18 estimate for CFC-11 emissions from eastern mainland China for 2014 – 2017 is 8.7 ± 1.0 Gg
19 yr⁻¹ higher than the population-weighted inventory-based estimate during this period⁷. When
20 run forward with our inversion-derived emission estimates, the models reproduce the
21 increasing mole fractions during pollution events at the monitoring sites (Extended Data
22 Figure 3). When using data from either Hateruma or Gosan, a rise in emissions is inferred for
23 eastern mainland China (Extended Data Figure 4). However, the magnitude of the rise is
24 smaller and more uncertain when Hateruma is used alone, likely because measurements at
25 this site are less sensitive to the major emissions sources. The derived emissions are relatively

1 insensitive to the magnitude or distribution of *a priori* emissions used in the Bayesian
2 inversion frameworks (Extended Data Figure 5). Emissions derived for western Japan and
3 both countries on the Korean peninsula are substantially smaller than those derived for
4 eastern mainland China, and emission magnitudes derived for Japan and Korea did not
5 change significantly during the study period (Extended Data Figure 6).

6
7 To provide context to these eastern Asian emission increases, we provide an update to the
8 global emission magnitudes^{3,4}. Long-term observations of baseline mole fractions, which are
9 representative of air masses that have not been substantially influenced by nearby sources,
10 are used to infer changes in the global emission rate of CFC-11^{3,4}. Using a two-dimensional
11 model of atmospheric transport and chemistry, and assuming a lifetime of 52 years for CFC-
12 11²⁰, we find that 64 ± 2 Gg yr⁻¹ were emitted on average between 2008 and 2012 when
13 observations from either AGAGE or NOAA were used (see Methods; Figure 2). These
14 emissions increased to a mean value of 75 ± 3 Gg yr⁻¹ or 80 ± 3 Gg yr⁻¹ for 2014 to 2017,
15 using AGAGE or NOAA data, respectively, which represents a rise of 11 ± 3 Gg yr⁻¹ (17 ± 5
16 %) or 17 ± 3 Gg yr⁻¹ (26 ± 5 %) above emissions during the 2008 – 2012 period. Our update
17 shows that the global emission increase previously reported through 2016^{3,4} has persisted
18 through 2017. Consistent with previous studies^{3,4}, our simplified global modelling approach
19 assumes that atmospheric transport did not vary interannually and our uncertainty estimates
20 do not account for the potential influence of interannual changes in dynamics on the derived
21 emissions. However, it has been demonstrated that changes in atmospheric transport may
22 have contributed to up to 50% of the slow-down in the rate of decline in global mole fraction,
23 at least between 2014 and 2016³. Therefore, this inferred average global emission increase,
24 and those that were previously reported, may over-estimate the true global emissions rise in
25 recent years.

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When compared to the AGAGE or NOAA-derived global emissions increase, our estimated rise for eastern mainland China accounts for $64 \pm 32 \%$ or $41 \pm 20 \%$ of the total, respectively. However, given that our global emissions increase may be over-estimated by up to a factor of two, owing to unaccounted-for changes in atmospheric dynamics³, the fraction of the global emission increase accounted for by eastern mainland China may actually be substantially higher. Hence, these results demonstrate that increased emissions from eastern mainland China since 2012 can account for a large fraction of the concurrent rise in global emissions. However, we cannot rule out additional, but probably smaller, increases from other regions of the world (e.g. northern, southern and western Asia, Africa and South America), potentially including other parts of China, to which our current measurement networks are insensitive.

The spatial distribution of emissions inferred within our eastern mainland China region for the periods 2008 – 2012 and 2014 – 2017 shows the largest change in or around the provinces of Shandong and Hebei, and to a lesser extent near Shanghai (Figure 3). This change is evident across all four inversion results (Extended Data Figure 7). Due to the effect of atmospheric mixing, and because we used a small number of monitoring stations that are relatively far from the major sources, it is not possible to discern the location of emissions sources at finer scales than provinces, or groups of provinces, in the north-eastern part of China. Therefore, it is unclear whether these results indicate that the new emissions originate from a small number of point sources or are more distributed within these regions.

A number of considerations suggest that the increase in emissions from China is unlikely to originate from increased rates of escape from pre-phase-out banks of CFC-11 in existing

1 foam or chillers. First, the spatial distribution of our derived emissions rise is unrelated to
2 population density (Figure 3), whereas the distribution of banks of foams should be roughly
3 distributed by population. Second, our cumulative emissions from eastern mainland China
4 between 2014 and 2017 (56 ± 7 Gg) are similar to an inventory-based estimate of the CFC-11
5 bank for all of China in 2013 (54 Gg)⁷, or almost three times the population-scaled bank for
6 this region, implying that the bank in 2013 was not large enough to accommodate the
7 emissions that occurred in subsequent years. Third, even given the possibility that the
8 Chinese bank size was underestimated for 2012, the emission increase derived for eastern
9 mainland China would require that the bank release fraction increase by around an order of
10 magnitude over the course of five years, a change that seems highly unlikely (**Error!**
11 **Reference source not found.**). To bring about such an increase in release rate from the bank
12 would require new emissions from the disposal and destruction of refrigerators more than ten
13 times higher than recently estimated for the whole of China between 2014 and 2017²¹, or a
14 larger and more rapid increase in emissions from the demolition of old buildings than was
15 previously predicted for the entire world over a 20-year period (2020 – 2040)²².

16

17 On the basis of this reasoning, it is likely that the new emissions of CFC-11 are the result of
18 new production that has not been reported to the UNEP Ozone Secretariat. Further
19 information is required to confirm which processes are responsible for releasing this newly
20 produced CFC-11 to the atmosphere. If consistent with historical usage, it would be expected
21 that emissions have primarily occurred during, or following, foam blowing, rather than
22 directly during production. Therefore, the regions where emissions have been identified in
23 this study are not necessarily the same as the locations where the compound has been
24 produced. It may be expected that new production of CFC-11 should be accompanied by new
25 emissions of carbon tetrachloride (CCl_4), the starting reagent for producing CFC-11, and

1 CFC-12 (CF₂Cl₂), which is typically co-produced with CFC-11. Whilst new emissions of
2 CCl₄ have been identified from parts of eastern China in recent years²³, we do not find
3 evidence for an increase in emissions of CFC-12 (Extended Data Figure 9). Given the
4 different historical uses of CFC-11 and CFC-12 (primarily in foam blowing and refrigeration,
5 respectively), the relationship between production and the subsequent rate of emission may
6 also be very different for these two substances. Therefore, the lack of a detectable increase in
7 CFC-12 emissions does not preclude a recent rise in CFC-11 production.

8

9 Our results show that new CFC-11 emissions from eastern mainland China explain a
10 substantial fraction of the post-2012 rise in global emissions, and are likely due to new,
11 unreported production and use. It is important to recall that before this period, global
12 emissions had not declined as expected since the early 2000s, and by 2012 were around 10
13 Gg yr⁻¹ higher than projections³. In contrast to the recent rise, we propose that emissions from
14 eastern mainland China are unlikely to have contributed significantly to this earlier “plateau”,
15 because our derived emissions for this region during 2008 – 2012 are relatively small (6.4 ±
16 1.2 Gg yr⁻¹) and consistent with estimated release rates from banks⁷. Other countries, and
17 indeed the rest of China, which are not well observed by the current monitoring networks
18 may also have contributed to the unexpected trends in CFC-11, which have now been
19 ongoing for almost two decades. Further investigation will be required to determine which
20 processes have led to the inferred recent increase in emissions from China (and potentially
21 elsewhere) and the magnitude of any associated, unreported CFC-11 production. Depending
22 on which processes are responsible, the new emissions may represent only a fraction of the
23 total new production, with the remainder contained in more slowly emitting banks (e.g.
24 foams). Ultimately, because nearly all known uses of CFC-11 are dispersive, it is the total

1 production magnitude that will determine the overall impact of these activities on the ozone
2 layer and its recovery.

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16

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15

16 **Author contributions**

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19 A.L.R., X.F. and S.H. carried out atmospheric model simulations and inverse analysis with
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23

24 **Additional information**

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3

4 **Methods**

5 **Data**

6 Observations were made at a frequency of around 34 measurements per day at the remote
7 AGAGE stations using gas chromatograph (GC) electron capture detection⁴. These stations
8 are at Mace Head, Ireland (53.3 N, 9.9 W), Trinidad Head, CA, USA (41.1 N, 124.2 W),
9 Ragged Point, Barbados (13.2 N, 59.4 W), Cape Matatula, American Samoa (14.2 S, 170.7
10 W) and Cape Grim, Tasmania, Australia (40.7 S, 144.7 E). At the AGAGE station at Gosan,
11 Republic of Korea (33.3 N, 126.2 E) around 12 measurements were made each day using
12 GC-mass spectrometry (MS)²⁴. Measurements at the NIES, AGAGE-affiliate station at
13 Hateruma, Japan (24.1 N, 123.8 E) were made at a frequency of around 20 measurements per
14 day using GC-MS²⁵. AGAGE measurements are reported on the Scripps Institution of
15 Oceanography (SIO) 2005 scale and NIES measurements are reported on the NIES-08 scale.
16 Hateruma data were converted to the SIO-05 scale by multiplying by an intercalibration
17 factor of 0.996, determined through exchange of air samples. The estimated repeatability of
18 the Hateruma data was approximately 0.8%, and the average repeatability of the Gosan data
19 was approximately 0.2%, although this varies throughout the time-series.

20

21 NOAA's background atmospheric CFC-11 measurements from three different methods were
22 combined into a single atmospheric history for this analysis (for the combined record, see
23 <ftp://ftp.cmdl.noaa.gov/hats/cfcs/cfc11/combined/> for data and
24 <https://www.esrl.noaa.gov/gmd/hats/combined/CFC11.html> for a description of the
25 methodology). Those methods include GC-ECD measurements at hourly frequency using on-

1 site instrumentation, and the GC-ECD and GC-MS (the latter included only since 2010)
2 analysis of glass and stainless-steel flasks collected approximately weekly at 12 locations
3 across the globe. Flasks were analysed in Boulder, CO, USA³. NOAA data are reported
4 using the NOAA-2016 scale.

5

6 Differences between the NOAA-2016 and the SIO-05 scales are 1% or less⁴. Therefore,
7 differences between the AGAGE and NOAA-derived global emissions (Figure 2) are not
8 strongly influenced by calibration differences. Global inversions were carried out using
9 AGAGE and NOAA data on their respective calibration scales. Uncertainties due
10 inaccuracies in the calibration scales were not included in the global emissions estimates (see
11 below).

12

13 **Regional atmospheric chemical transport models**

14 We use two backward-running Lagrangian chemical transport models to estimate the
15 sensitivity of the atmospheric observations to fluxes in the eastern Asian region: NAME¹⁴
16 and FLEXPART¹⁵. Both models simulate transport of CFC-11 by advection and random
17 diffusion, by atmospheric turbulence, of hypothetical particles of the gas. The “footprint” of
18 each measurement, calculated from these simulations, describes the sensitivity of the
19 observation to emissions from surrounding model grid cells (Extended Data Figure 2). A
20 comparison of the simulated and observed data is presented in Extended Data Figure 3.

21

22 NAME was driven by meteorological fields from the UK Met Office Unified Model²⁶, the
23 spatial resolution of which increased in the study period from 0.563 to 0.141° longitude and
24 0.375 to 0.094° latitude, whilst the temporal resolution has remained fixed at 3 hours. 20,000
25 particles per hour were transported backward in time for 30 days in NAME in a domain

1 bounded at 5°S and 74°N and 55°E and 192°E. When particles are within the lowest 40 m of
2 the atmosphere, it was assumed that they interacted with the surface and could intercept
3 emissions sources^{17,23}. The NAME-HB framework uses a climatological *a priori* estimate of
4 the mole fraction at the boundary of the domain from the NASA 3D GEOS Chemistry
5 Climate Model²⁷. In contrast, the NAME-InTEM inversion estimated the baseline mole
6 fraction at each measurement station (see below).

7

8 FLEXPART was run in different modes by the MIT and Empa groups. FLEXPART-Empa
9 released 50,000 particles in each 3-hour interval and traced them backwards in time for 10
10 days, driven by operational ECMWF analysis with 1° x 1° global resolution and 0.2° x 0.2°
11 resolution for north-eastern China (105°E to 125°E and 30°N to 50°N). FLEXPART-MIT
12 released 40,000 particles in each 3-hour interval and traced them backwards in time for 20
13 days in a global domain, driven by operational ECMWF analysis at 1° x 1° global resolution.

14

15 **Regional inversion frameworks**

16 Fluxes were derived using four different Bayesian inversion frameworks. Bayesian inverse
17 modelling systems use an *a priori* estimate of emissions, which, when combined with
18 chemical transport models, can be subsequently updated to bring the models into consistency
19 with observations. In our study, different *a priori* estimates were used in each regional
20 inversion, although in every case these estimates primarily served to weakly limit the spatial
21 area from which emissions were inferred (they were limited to being over land, but not
22 further constrained to any particular land-based regions), and to provide the inversion with an
23 approximate magnitude for the fluxes (by typically being some fraction of the global total).
24 The magnitude of the *a priori* emissions was constant for each year of the study, so that any
25 temporal variation in the *a posteriori* emissions was driven only by the data. Bayesian

1 frameworks require that some uncertainty is placed on these prior estimates along with an
2 estimate of the model-measurement uncertainty covariance (see below). The hierarchical
3 approach allows these uncertainty factors, along with other parameters, to be estimated in the
4 inversion²⁸. Our inversions were shown to be relatively insensitive to the choice of prior
5 emissions magnitude or distribution (Extended Data Figure 5).

6
7 The trans-dimensional hierarchical Bayesian inversion used with NAME footprints (NAME-
8 HB) estimates the model-measurement uncertainty in the inversion²⁸. Furthermore, the level
9 of spatial disaggregation of the emissions field can also be explored¹⁶. This scheme is solved
10 using reversible-jump Markov chain Monte Carlo (rj-MCMC)²⁹. The inversion is split into an
11 inner domain (74°W to 160°W, 22°N to 50°N), where spatial disaggregation can occur, and
12 an outer domain split into four fixed regions to the North, South, East and West extending to
13 the boundary of the NAME domain. Emissions are estimated at variable resolution
14 (determined by the rj-MCMC algorithm) in the inner domain and from the four fixed regions
15 outside this domain. Simultaneously, boundary conditions at the four domain edges were
16 solved for by scaling the *a priori* mole fraction fields at these edges. Our *a priori* estimate of
17 CFC-11 emissions were 20 Gg yr⁻¹ distributed uniformly over land (not water) in the inner
18 domain and a further 20 Gg yr⁻¹ in the outer domain, meaning that the combined domain
19 accounts for roughly two thirds of global emissions. This results in national *a priori*
20 emissions estimates of 17.3 Gg yr⁻¹ for China (5.5 Gg yr⁻¹ for eastern mainland China) and
21 1.2, 0.31 and 0.42 Gg yr⁻¹ for Japan, Republic of Korea and D. P. R. Korea, respectively.
22 Within each country, these prior fluxes were uniformly distributed in space. The uncertainty
23 on these flux estimates was assumed to be lognormally distributed, with a mode fixed to the *a*
24 *priori* estimated emission and a logarithmic standard deviation of 10, defined as the square-
25 root of the ratio of the exponential of the mean to the mode. Model uncertainties are informed

1 by the data in the hierarchical framework. The model error was assumed to be normally
2 distributed with zero mean and a standard deviation that was allowed to vary, with equal
3 probability, between 1 and 20 ppt, with an autocorrelation timescale between 1 and 120
4 hours. The measurement data and footprints were averaged into daily values, with a fixed
5 measurement uncertainty equal to the repeatability of the raw measurements plus the
6 variability of the measurements within the daily averaging period (a measure of the sub-daily
7 fluctuations that would not be resolved by the inversion). Emissions were assumed constant
8 throughout the year, and emissions and monthly boundary condition scaling factors were
9 estimated independently within each year.

10

11 Output from NAME was also used with the InTEM inversion framework (NAME-InTEM)¹⁷.

12 *A priori* CFC-11 estimates were given large uncertainties, namely 22 (0-147) Gg yr⁻¹ for
13 China, 5 (0-35) Gg yr⁻¹ for eastern mainland China, and 2 (0-20), 0.3 (0-3.2) and 0.4 (0-3.8)
14 Gg yr⁻¹ for Japan, Republic of Korea and D. P. R. Korea, respectively. This is based on 50
15 Gg yr⁻¹ uniformly emitted across the whole eastern Asian land area. Observational
16 uncertainty was time varying and was estimated as the variability of the observations in a six-
17 hour moving window plus the measurement repeatability determined from repeat
18 measurements of the on-site calibration standards. Model uncertainty was estimated every
19 two hours as the larger of the median of all pollution events at each station in a year or 16.5%
20 of the magnitude of the pollution event. A temporal correlation of 12 hours was assumed in
21 the model uncertainty at each station. An analytical solution was found that minimized the
22 residual between the model and the observations and the difference between the posterior and
23 prior flux estimate, balanced by the uncertainties of both. The mole fraction baseline at each
24 station was estimated in the inversion following Arnold et al. (2018)¹⁷. The variable
25 resolution of the inversion grid was calculated and refined within InTEM based on the

1 magnitude of the footprint and emissions from each grid box. The inversions were run
2 multiple (24) times per year, each time with a randomly generated sub-sample (90%) of the
3 available observations (10% removed in 5-day blocks), to further explore the uncertainty.
4 Emissions and uncertainties were averaged across the 24 individual inversions thereby
5 assuming 100% correlation between uncertainties in these separate inversions.
6
7 The FLEXPART-MIT inversion derived fluxes using FLEXPART with an analytical
8 Bayesian framework³⁰. Prior flux fields were set to be spatially uniform over continental
9 eastern Asia. Emissions were solved within variable-resolution grid cells with the finest grid
10 ($1^\circ \times 1^\circ$) in eastern China and other eastern Asian countries³¹. *A priori* estimates of CFC-11
11 flux were 14.5 Gg yr^{-1} for China (1.5 Gg yr^{-1} for eastern mainland China) and 1.2, 0.27 and
12 0.27 Gg yr^{-1} for Japan, Republic of Korea and D. P. R. Korea, respectively. The uncertainty
13 on each grid cell was arbitrarily set to 1000% and the spatial correlation length was assumed
14 300 km, so that the total *a priori* emission uncertainty for China, for example, was 43.2 Gg
15 yr^{-1} . Baseline mole fractions were estimated at each site every 7 days³¹. Model-measurement
16 uncertainty for each 24-hour-averaged observation was estimated as the quadratic sum of 1%
17 of the baseline value (as a measure of baseline uncertainty), the measurement repeatability
18 and the standard deviation of the 24-hourly variability (as a measure of the model-data
19 “mismatch” uncertainty). Similarly to InTEM, *a posteriori* fluxes were derived using an
20 analytical approach that minimises a cost function that weights the measurement-model
21 residuals and the *a priori* estimate by their respective uncertainties.
22
23 Fluxes were derived with a second FLEXPART-based approach but using the FLEXPART-
24 Empa simulations and an alternative analytical Bayesian framework¹⁹. Prior fluxes were set
25 to a homogeneous value over all land areas with total Chinese and eastern mainland Chinese

1 emissions of 14.9 Gg yr⁻¹ and 2.7 Gg yr⁻¹, respectively, (0.66, 0.18, 0.22 Gg yr⁻¹ for Japan,
2 Republic of Korea and D. P. R. Korea). Observations were aggregated to 3-hourly averages
3 to s gridded annual mean emissions. The baseline was optimized as part of the state vector,
4 independently for both sites. No *a priori* covariance between emissions and baseline were
5 assigned. Parameters describing the covariance structure of the data-mismatch uncertainty
6 matrix and the *a priori* uncertainty matrix were estimated using a maximum likelihood
7 approach¹⁹. These parameters include an estimate of the model uncertainty at the individual
8 sites, the *a priori* baseline uncertainty and its temporal correlation length scale, a domain-
9 total *a priori* uncertainty, and a spatial correlation length scale. The domain-total *a priori*
10 uncertainty was estimated to range from ±45 % (in 2008) to ±125 % (in 2016), which
11 translates to a regional *a priori* uncertainty of ±100 % to ±270 % for the eastern mainland
12 China region.

13
14 Where the emissions, derived from multiple regional inversions and multiple years, were
15 combined, we took the mean of the *a posteriori* annual inversion means. Mean emissions are
16 calculated throughout the text for the periods 2008 – 2012 and 2014 – 2017 (inclusive). The
17 year 2008 was used to bound the first period, when a whole year of Gosan data first became
18 available. A previous study³, which estimated global emissions using NOAA data, omitted
19 the year 2013, because global emissions in this year appeared to be intermediate between two
20 periods of more constant emission. Whilst our estimates, which use multiple models and
21 measurement networks, are less clear on whether such an intermediate period occurred, we
22 also omit the year 2013, so that our estimates can be more directly compared with this
23 previous work. Uncertainties in the multi-inversion means were calculated from the *a*
24 *posteriori* uncertainty from the individual models and years, assuming that these uncertainties
25 were fully correlated (i.e. assuming that the different inversions did not provide statistically

1 independent estimates of emissions). The latter assumption attempts to account for the fact
2 that the inversions share the same models, which will suffer from similar systematic
3 uncertainties (in the case of the two NAME inversions or two FLEXPART inversions). Such
4 systematic uncertainties are not explored in any of the inversion frameworks and are
5 therefore not reflected in the individual *a posteriori* emission estimates. Therefore, the
6 individual uncertainty estimates are likely to be underestimates of the full uncertainty (as
7 seen by the non-overlapping uncertainties in some years in, for example, Figure 2). It is also
8 possible that, for the same reason, our multi-inversion uncertainty is an under-estimate.
9
10 Based on a consideration of the sensitivity of the observations to potential sources
11 surrounding the measurement sites (note that the NOAA and AGAGE network currently have
12 a limited number of stations which are close enough to industrialised regions for the detection
13 of regional emission sources), we have opted to focus on the total emissions rise from a
14 region that we denote eastern mainland China (Extended Data Figure 2, a similar sub-
15 division was also used for western Japan). Further away from this region, atmospheric mixing
16 of plumes that emanate from emissions sources makes their impact on the observations small,
17 compared to measurement and modelling uncertainty (which is why the existing, sparse
18 network is insensitive to large parts of the globe). The choice of which Chinese provinces to
19 include in this region was somewhat subjective and could alter the derived emissions rise and
20 its uncertainty. Inclusion of the provinces adjacent to our eastern mainland China region
21 (excluding the very large Inner Mongolia) would lead to an increase of approximately 15% in
22 the emissions rise derived between 2008-2012 and 2014-2017. However, the associated
23 uncertainty increased by around 30%. We note that the use of a larger region would explain a
24 larger (but more uncertain) fraction of the global emissions rise than obtained from the
25 eastern mainland China region alone.

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Global average emissions estimation

Global emissions were estimated from AGAGE or NOAA data with a least-squares method used previously for global CFC-11 emissions estimation³. Monthly mean baseline-filtered observations³² from the five remote AGAGE stations or the NOAA flask and *in situ* sites were averaged into semi-hemispheres, separated at the equator and 30° north or south (NOAA data were weighted by the sine of their latitude within each semi-hemisphere). Mole fractions were simulated for each semi-hemisphere using a 12-box model of the atmosphere with vertical divisions at 500 hPa and 200 hPa³³. Seasonally varying, inter-annually repeating advection and diffusion parameters were used to transport CFC-11 between model boxes. Loss was assumed only to occur in the stratospheric boxes, at a rate that was adjusted to provide a global mean steady-state lifetime of 52 years²⁰. Uncertainties related to the model's ability to simulate the data consisted of the sum of two factors. The first term, equal to the baseline variability in each month determined by the AGAGE network, was assumed to be a measure of the temporal representation error between the model and the data. A second fixed uncertainty (0.3%), equal to the approximate level of disagreement between the AGAGE and NOAA monthly semi-hemispheric means, was used to parameterize spatial and temporal differences in the sampling between the networks and uncertainties in the propagation of calibration scales to the sites. These uncertainties are propagated through to the emissions estimates. However, our estimates do not include an uncertainty contribution due to calibration scale uncertainty or atmospheric lifetime uncertainty³³. These terms are important for quantifying the absolute derived emissions values but have little influence on the change in emissions between time periods, which is the primary focus of this paper. Similarly to previous studies^{3,4}, our uncertainties do not include a contribution due to the assumption of

1 inter-annually repeating dynamics. To estimate the implications of this assumption would
2 require three-dimensional global model simulations, which were not available for this work.

3

4 **Data Availability**

5 Data from remote AGAGE stations and Gosan data are available from the AGAGE website
6 (agage.mit.edu). Hateruma data are available at World Data Center for Greenhouse Gases
7 (<http://ds.data.jma.go.jp/gmd/wdcgg/wdcgg.html>). NOAA data are taken as the “combined
8 set” data record (a best-estimate record based on flask samples and in-situ measurements),
9 available from the NOAA Global Monitoring Division data server
10 (<https://www.esrl.noaa.gov/gmd/dv/ftpdata.html>, or more specifically
11 <ftp://ftp.cmdl.noaa.gov/hats/cfcs/cfc11/combined/>).

12

13 **Code availability:**

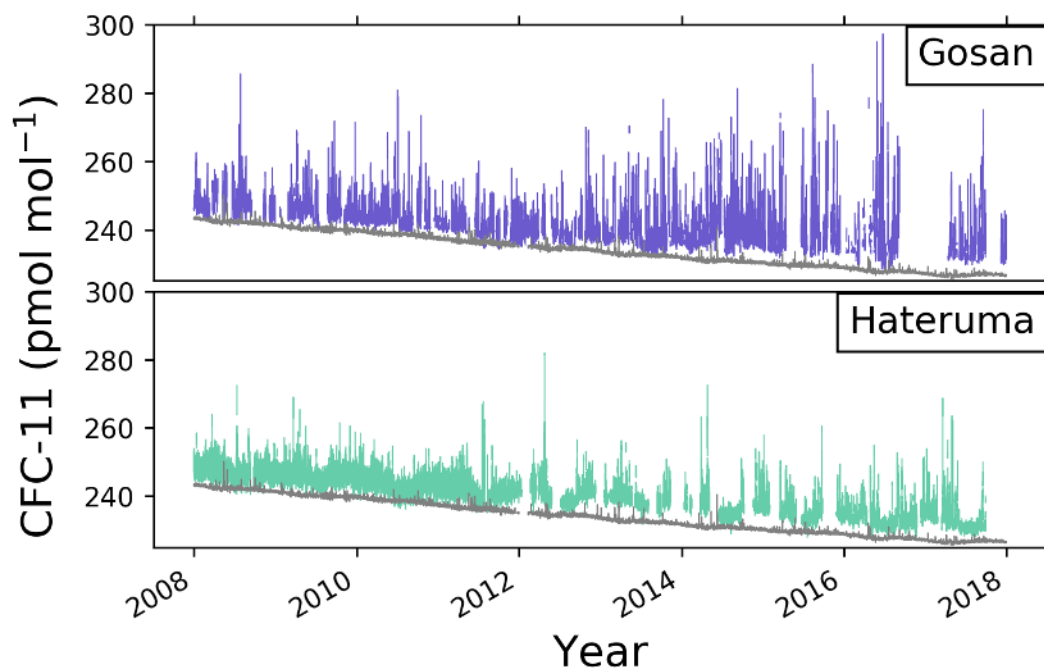
14 The inversion models of NAME-HB, NAME-InTEM, FLEXPART-MIT and FLEXPART-
15 Empa are used to determine the regional CFC-11 emissions. Enquiries about the model codes
16 should be directed to M.R., A.J.M., S.R. and R.G.P. Licences to use NAME and InTEM are
17 available for research purposes via a request to the UK Met Office or on request from A.J.M.
18 and A.L.R... The code for the NAME-based hierarchical Bayesian inversion (NAME-HB) is
19 available on request from M.R. and L.M.W.. The code of the dispersion model FLEXPART
20 is available from www.flexpart.eu. The code for the FLEXPART-based Bayesian inversion
21 (FLEXPART-MIT) is available on request from X.F.. The inversion code used by Empa is
22 available from <https://doi.org/10.5281/zenodo.1194642> or on request from S.H..

23

24

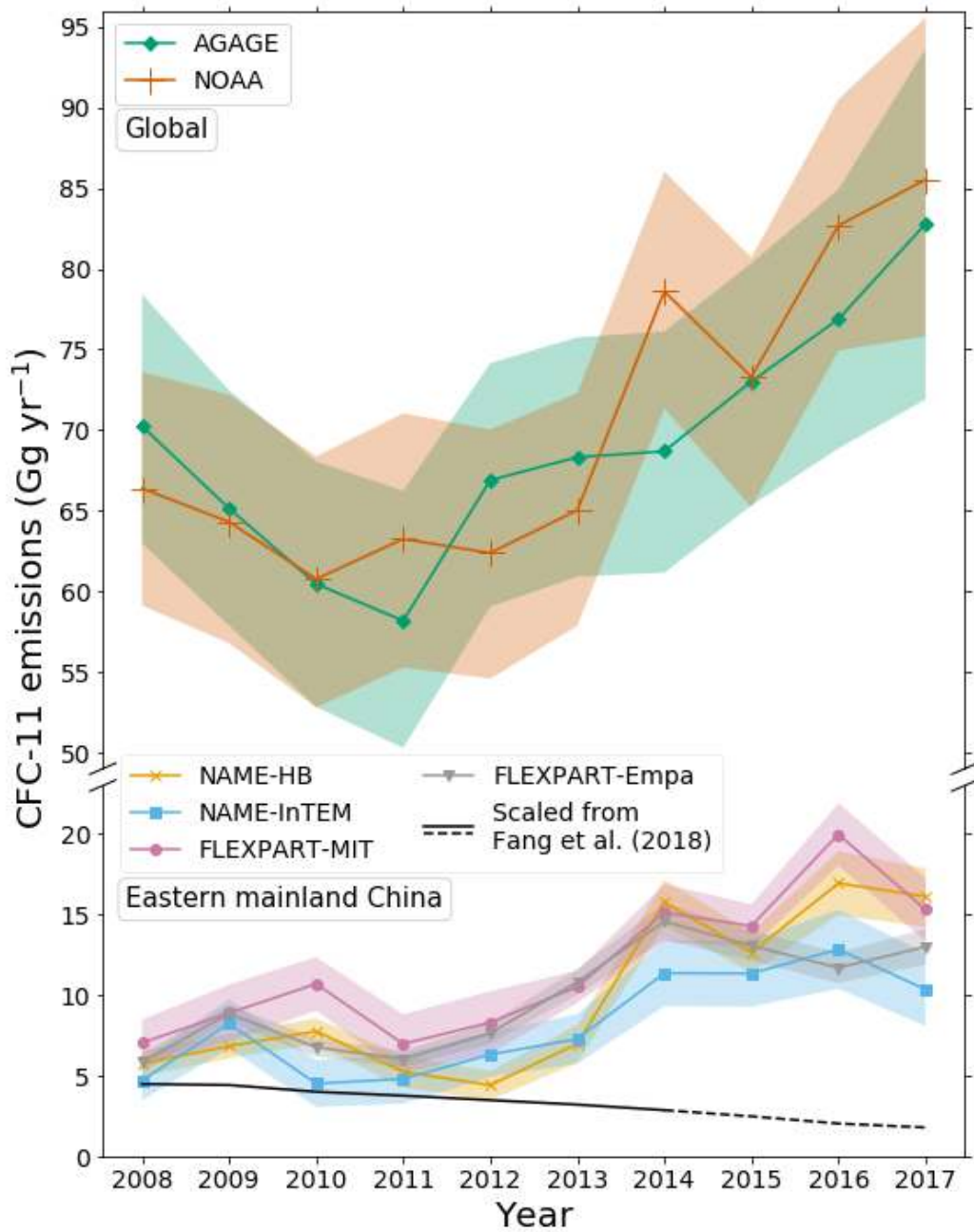
1 **Figures**

2



3

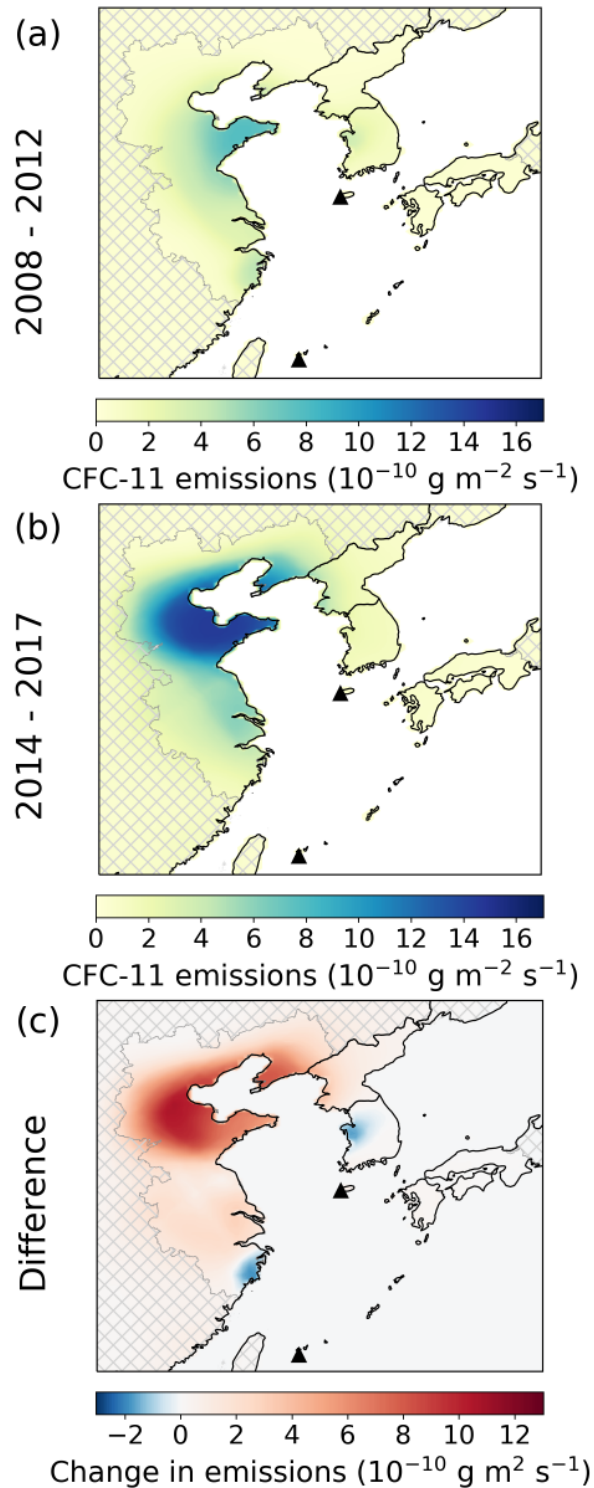
4 Figure 1. Dry-air mole fraction observations for CFC-11 at Gosan, Jeju Island, Republic of
5 Korea (33.3 °N, 126.2 °E) and Hateruma, Japan (24.1 °N, 123.8 °E). The grey lines show the
6 mole fraction observations at Cape Grim, Tasmania, Australia (40.7 °S, 144.7 °E) for
7 comparison.



1

2 Figure 2. CFC-11 emissions derived from atmospheric observations. The upper part of the
 3 figure shows global emissions estimates derived using a global box model and AGAGE
 4 (green diamonds) and NOAA (orange crosses) mole fraction observations of baseline air. The
 5 lower part of the figure shows emissions estimates derived for eastern mainland China using
 6 measurements from Gosan (AGAGE, Republic of Korea) and Hateruma (NIES, Japan) and
 7 four inverse modelling frameworks: NAME-HB (yellow crosses), NAME-InTEM (blue

1 squares), FLEXPART-MIT (pink circles) and FLEXPART-Empa (grey triangles). Inventory-
2 based estimates for CFC-11 emissions from eastern mainland China (determined as total
3 Chinese emissions scaled by the fraction of the population, 35 %, that reside in that part of
4 the domain) are shown as a solid black line and a projection of the inventory estimates are
5 shown as a dashed line⁷. All lines and symbols represent the *a posteriori* mean, and the
6 shading, the 1-standard deviation uncertainty. Global emissions are higher than in a previous
7 study using NOAA data³, because a shorter atmospheric lifetime was used here²⁰.



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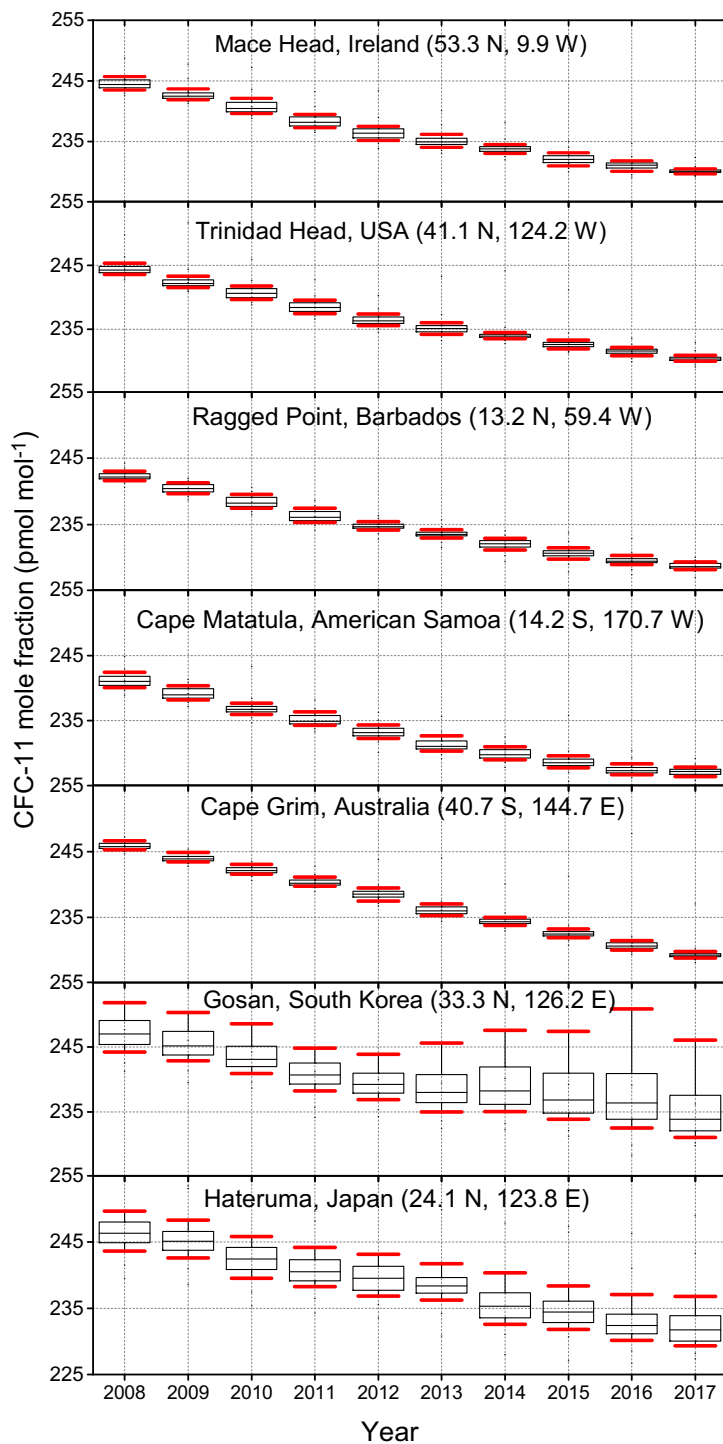
2 Figure 3. Spatial distribution of the derived CFC-11 fluxes in the NAME-HB inversion. a)
 3 mean emissions for 2008 – 2012; b) mean emissions 2014 – 2017; c) Change in emissions
 4 from 2008 – 2012 to 2014 – 2017. Equivalent plots for the other inversions are shown in
 5 Extended Data Figure 7. The hatched areas indicate regions of the domain to which the

1 observations have low sensitivity (Extended Data Figure 2), and therefore, from which the
2 derived emissions have high uncertainty. As a result, only emission magnitudes and emission
3 changes for the non-hatched regions are included in the values quoted in the main text.

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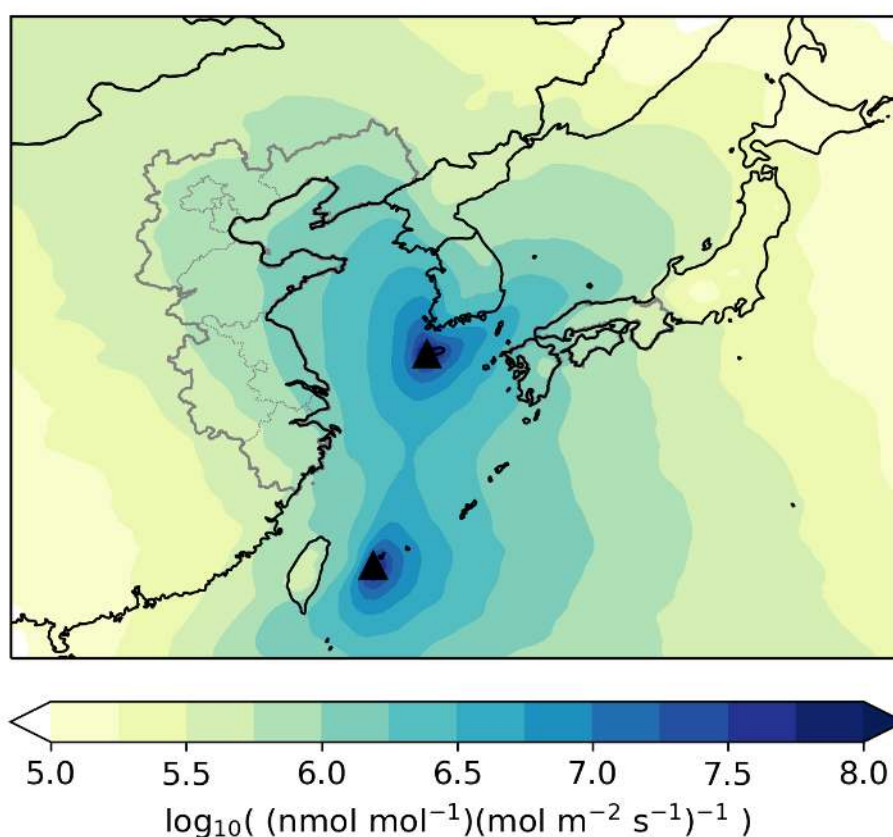
1 Extended data



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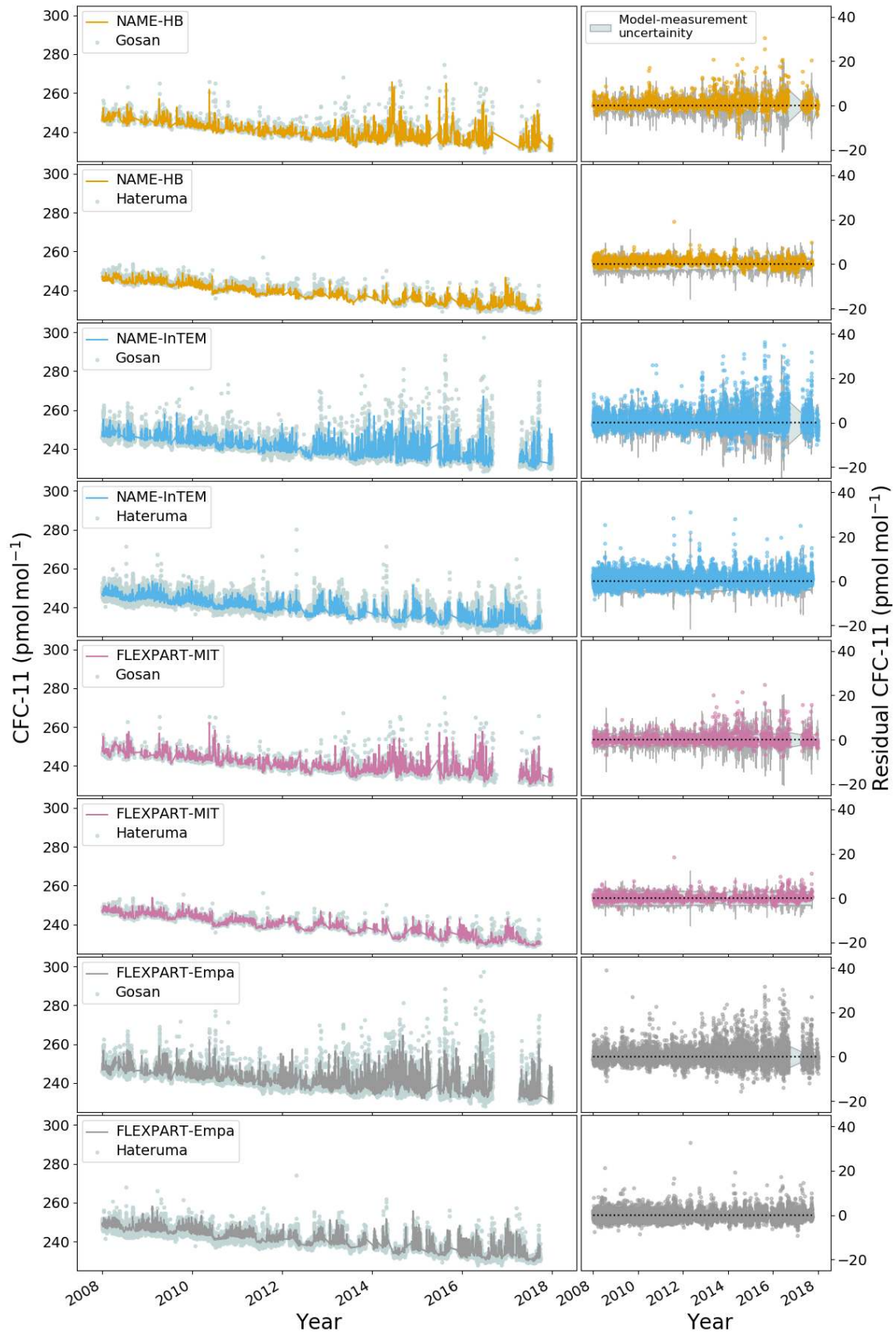
3 Extended Data Figure 1. Box and whisker plots of mole fractions measured at different sites
4 in the AGAGE network and at affiliated AGAGE sites. Boxes indicate the 25th and 75th
5 percentiles of the individual measurement data (approximately 2-hourly), with the median

1 shown as a line within each box. The whiskers show the 10th and 90th percentiles. The lower
2 percentiles are typically representative of baseline (“unpolluted”) mole fractions, and the
3 difference between the lower and higher percentiles indicates the magnitude of the above-
4 baseline events due to the interception of air masses containing recently emitted CFC-11.
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6
7 Extended Data Figure 2. Mean sensitivity of observations at Gosan (33.3 N, 126.2 E) and
8 Hateruma (24.1 N, 123.8 E), indicated by the black triangles, to emissions from 2008 to 2017
9 from the NAME model simulation. Thin grey lines show the boundaries of the provinces
10 within the region we denote “eastern mainland China”. This region contains the provinces

- 1 Anhui, Beijing, Hebei, Jiangsu, Liaoning, Shandong, Shanghai, Tianjin and Zhejiang. The
- 2 west of the grey line transecting Japan, containing the regions Chūgoku, Kansai, Kyūshū &
- 3 Okinawa, and Shikoku, is the region we denote “western Japan”.



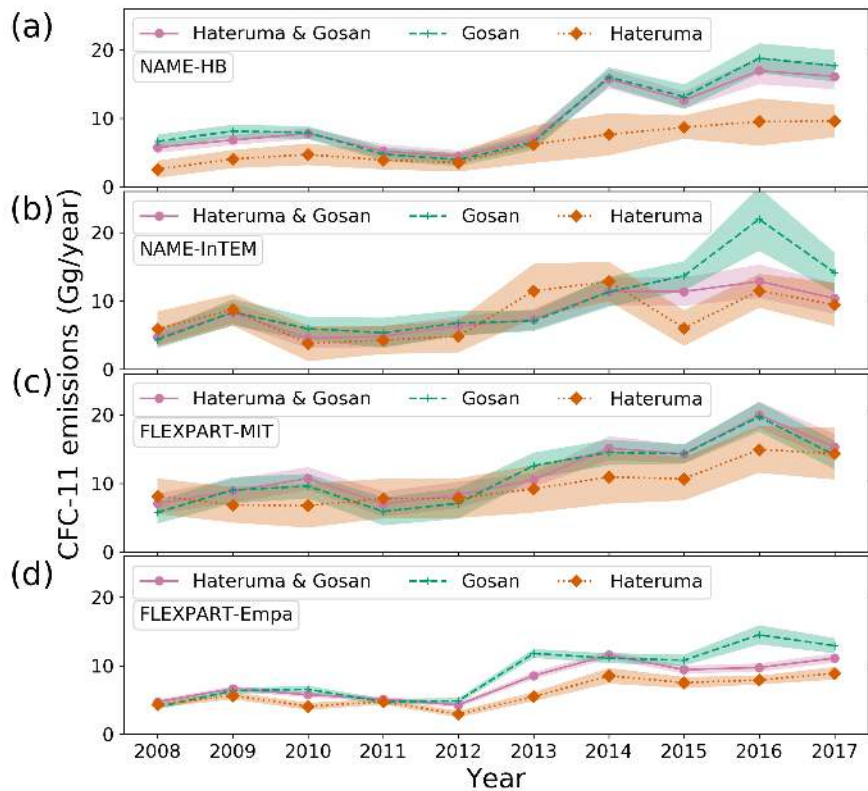
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2 Extended Data Figure 3. Left hand panel: A comparison of simulated and observed mole

3 fractions at Gosan and Hateruma from the four inversion frameworks. Right hand panel:

1 Residuals between the simulated and observed mole fractions (data minus model). One
 2 standard deviation model-data mismatch uncertainties assumed in the inversions are indicated
 3 by shading. Simulated mole fractions are derived from the *a posteriori* emissions. For the
 4 NAME-InTEM, FLEXPART-EMPA, NAME-HB and FLEXPART-MIT inversions, 2-
 5 hourly, 3-hourly, 24-hourly and 24-hourly averaging was applied to the model and data,
 6 respectively.

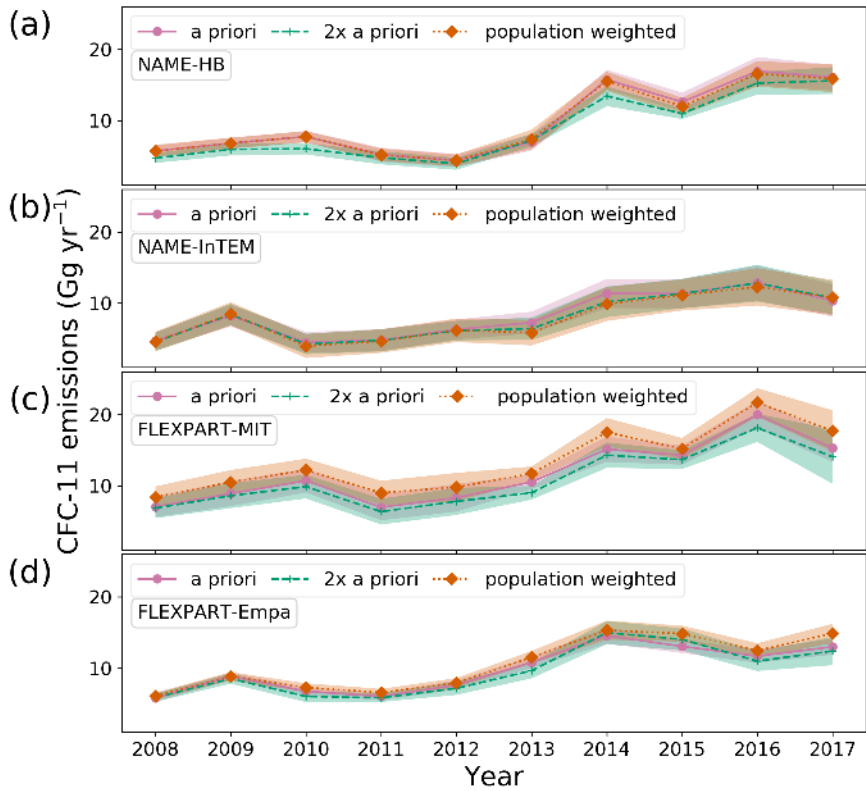
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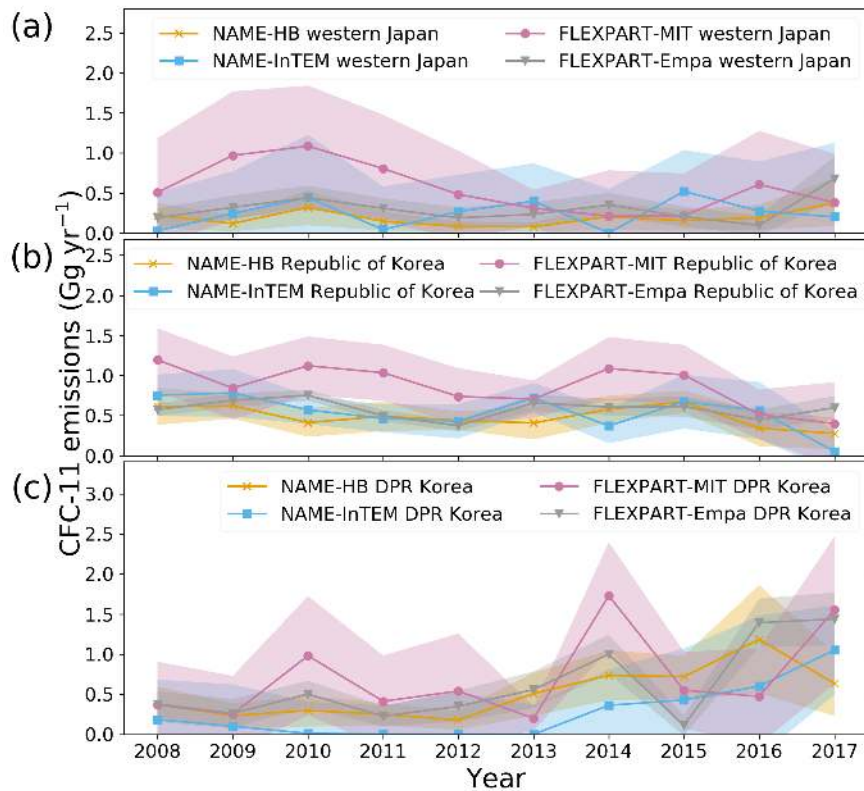
9 Extended Data Figure 4. Inverse emissions estimates for eastern mainland China derived
 10 using only the Gosan CFC-11 measurements (green dashed line with crosses), only the
 11 Hateruma (red dotted line with diamonds) CFC-11 measurements, or using both
 12 measurement records in the inversion analysis (pink solid line with circles). The estimates

- 1 shown use the (a) NAME-HB, (b) NAME-InTEM, (c) FLEXPART-MIT and (d)
- 2 FLEXPART-Empa inversion techniques.



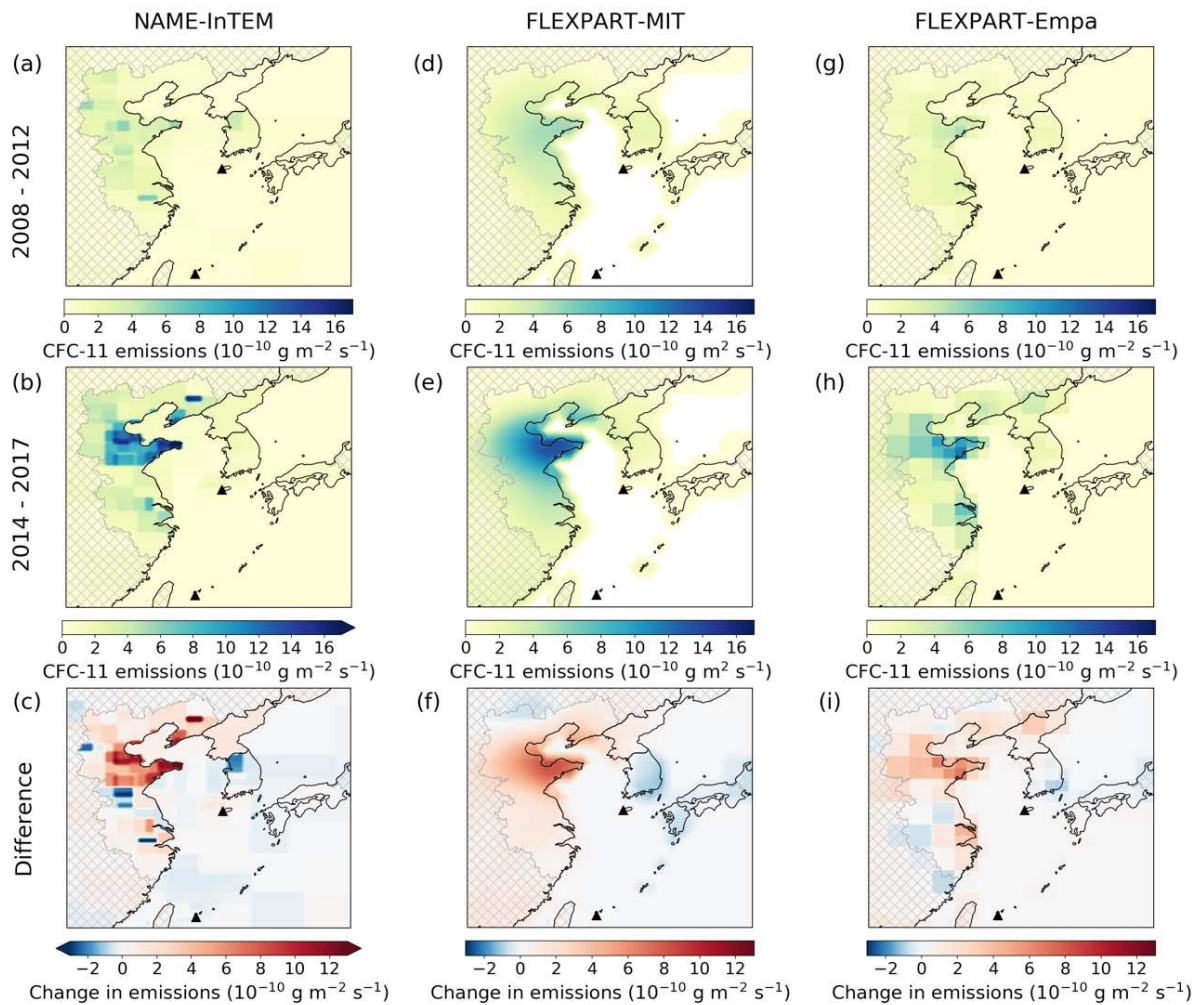
- 3
- 4 Extended Data Figure 5. Derived emissions for eastern mainland China for different *a priori*
- 5 emission magnitudes and different spatial distributions. The inversions are as described in
- 6 Methods (“*a priori*” – pink solid with circles), with *a priori* emissions twice as high (“2x
- 7 *a priori*” – green dashed line with crosses), or the same magnitude but distributed in space
- 8 according to population density (“population weighted” – red dotted line with diamonds).
- 9 Figures show the estimates using the (a) NAME-HB, (b) NAME-InTEM, (c) FLEXPART-
- 10 MIT and (d) FLEXPART-Empa inversion methods.

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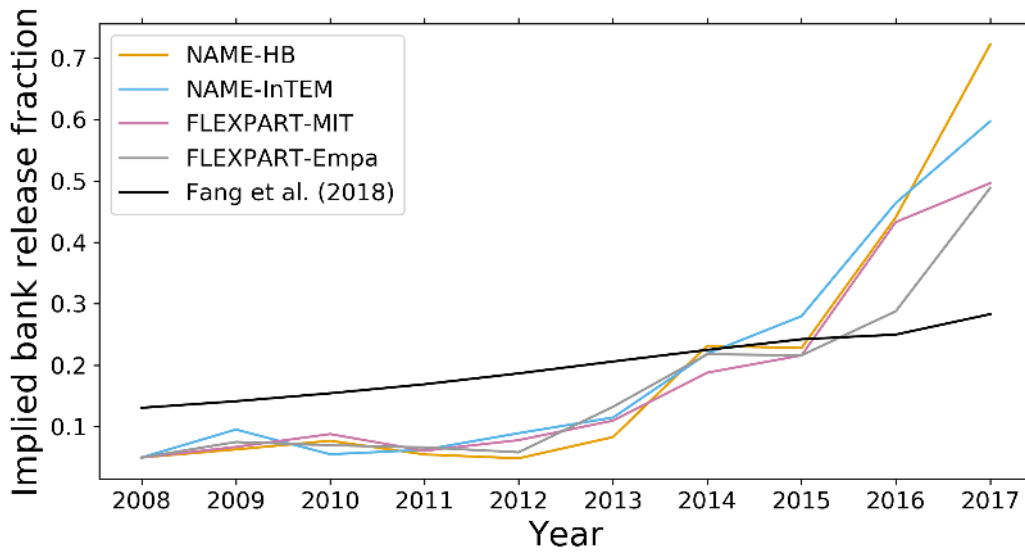
2 Extended Data Figure 6. Top-down emissions estimates for (a) western Japan, (b) the
 3 Republic of Korea and (c) the Democratic People’s Republic of Korea using the NAME-HB
 4 (yellow exes), NAME-InTEM (blue squares), FLEXPART-MIT (pink circles) and
 5 FLEXPART-Empa (grey inverted triangles) inverse frameworks described in Methods. See
 6 Extended Data Figure 2 for the definition of the western Japan region. All lines and symbols
 7 are the *a posteriori* mean and the shading shows the one standard deviation uncertainty.



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Extended Data Figure 7. Mean CFC-11 emission fluxes and difference maps, similar to Figure 3, for NAME-InTEM (a-c), FLEXPART-MIT (d-f) and FLEXPART-Empa (g-i) using the inversion framework described in the Methods. Figures (a), (d) and (g) show the average spatial emissions for the period 2008-2012; (b), (e) and (h) show the average spatial emissions for the period 2014-2017; and (c), (f) and (i) show the difference in emissions from the 2008-2012 period to the 2013-2017 period, using NAME-InTEM, FLEXPART-MIT and FLEXPART-Empa respectively.

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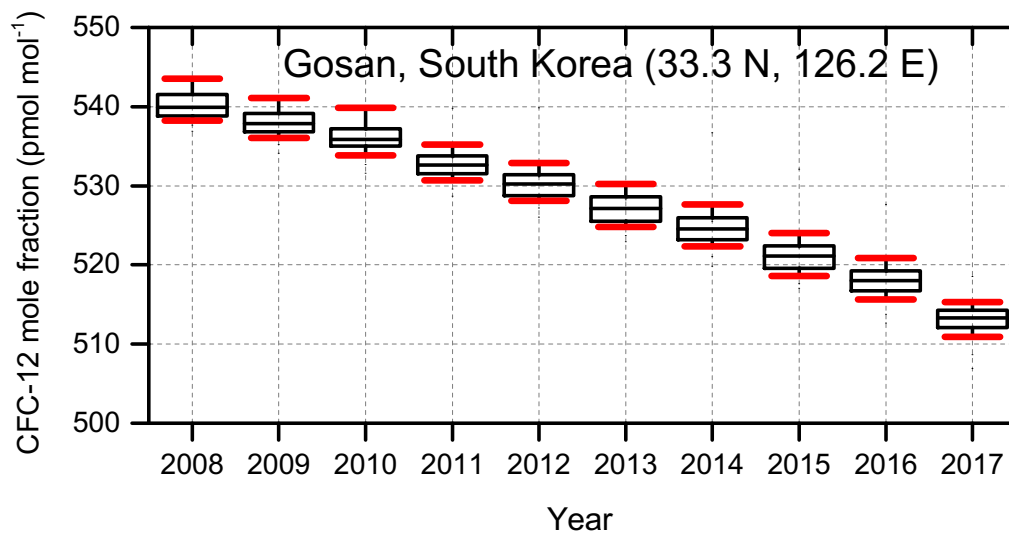
3 Extended Data Figure 8. The implied bank release fraction of CFC-11 from the eastern
4 mainland China region defined in Extended Data Figure 2, if there is no new production. This
5 calculation assumes that the 2008 emissions estimate are 5% of the bank size (estimated to be
6 toward the upper limit of expected bank fractional release rate³). The coloured lines show the
7 implied bank release fraction for each year after 2008, required to sustain the mean a
8 *posteriori* emissions from the NAME-HB (yellow), NAME-InTEM (blue), FLEXPART-MIT
9 (pink) and FLEXPART-Empa (grey) inversions. The black line shows the bank release
10 fraction from a bottom-up study for the whole of China⁷.

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2 Extended Data Figure 9. Box and whisker plot of CFC-12 mole fraction at the Gosan
 3 measurement site. Boxes indicate the 25th and 75th percentiles of the (approximately 2-
 4 hourly) measurement data in a given year, and the median mole fraction is shown by the line
 5 inside each box. The whiskers show the 10th and 90th percentiles. The lower percentiles are
 6 typically representative of baseline mole fractions, and the difference between the lower and
 7 higher percentiles indicates the magnitude of the above-baseline events due to the
 8 interception of air masses containing recently emitted CFC-12.

9

10