

Perfluorocyclobutane (PFC-318, *c*-C₄F₈) in the global atmosphere

Mühle et al., acp-2019-267

Supplemental text, figures, and tables

AGAGE in situ data are available at

<http://agage.mit.edu/data>

<http://cdiac.ess-dive.lbl.gov/ndps/alegagage.html>

CSIRO and Bristol inversion results, firm data, etc. are given in

Mühle et al. *c*-C₄F₈ acp-2019-267 Supplemental tables.xlsx

Details on bottom-up emission inventories (UNFCCC, EDGAR, NIRs, WSC) for *c*-C₄F₈

Amongst the countries reporting to the United Nations Framework Convention on Climate Change (UNFCCC) (2016), a few countries report *c*-C₄F₈ emissions, most notably France, the USA, and Russia, and the global total ranges from 7 (1993) to 26 (2011) t yr⁻¹ (0.007–0.026 Gg yr⁻¹, 1 t = 1 metric ton = 1 tonne = 0.001 Gg). Several countries also or exclusively report emissions of an unspecified mix of PFCs as a sum of CO₂-equivalent (CO₂-eq.) emissions (using global warming potentials, GWP), which may contain *c*-C₄F₈ emissions, most notably Japan (3,260 (2013) – 19,900 (1997) Gg CO₂-eq.), followed by much smaller amounts from France (~139 (2000) – 518 (2012) Gg CO₂-eq.) and a few other European countries.

Based on the National Inventory Report (NIR) for France, their reported emissions of unspecified mix of PFCs, all from category 2G2 (SF₆ and PFCs from other product use), does not contain any *c*-C₄F₈.

Based on the NIR for the Netherlands and Austria, category 2E1 (integrated circuit (IC) or semiconductor (SC) production) could contribute a few to a few ten t yr⁻¹ of *c*-C₄F₈ per country if all emissions of unspecified mix of PFCs were *c*-C₄F₈, which is very unlikely. More likely seems that the GWP weighted mix of fugitive emissions from category 2E1 from these countries is similar to the mix for category 2E1 from European countries which report individual emissions for category 2E1. For the European Union, *c*-C₄F₈ represents ~0–8 % of the GWP weighted PFC mix (CF₄, C₂F₆, C₃F₈, *c*-C₄F₈) from 2E1. If the Netherlands and Austria emit 8 % of GWP weighted mix of PFCs in category 2E1 as *c*-C₄F₈, this would sum up to at most a few t yr⁻¹.

The United Kingdom NIR details that their unspecified PFC mix emissions are all from category 2B9 (fluorochemical production) and refers to the UK environmental agency's pollution inventory (<https://data.gov.uk/dataset/pollution-inventory>). Judging from this inventory and our knowledge of the listed PFC sources, they most likely do not emit any *c*-C₄F₈ and thus category 2B9 does not contain any *c*-C₄F₈ emissions for the UK.

The Japanese NIR details that their emissions of unspecified mix of PFCs stems from categories 2B9, 2E (SC, liquid crystals, and photovoltaic production), and 2F5 (solvents use). For Japan, category 2F5 is comprised of C₃F₁₄ and

C₆F₁₆ emissions, but no *c*-C₄F₈. *c*-C₄F₈ emissions from category 2E can be estimated using purchased amounts of *c*-C₄F₈ from the NIR and IPCC emission estimation methods (IPCC, 2006) and are likely at most a few t yr⁻¹. If all of Japan's emissions from category 2B9 (fugitive emissions from fluorochemical production) were *c*-C₄F₈, this could equate to several tens to two hundred t yr⁻¹. However, more likely is that the PFC emissions from category 2B9 are due to fugitive emissions from PFC production in Japan, and that their mix is similar to the PFC mix used in Japan for the electronics industries (2E) of CF₄, C₂F₆, C₃F₈, and *c*-C₄F₈. As detailed in the NIR, *c*-C₄F₈ used in category 2E represents 0–7 % of the total PFC mix (CO₂-eq.), which would equate to a few t yr⁻¹ from category 2B9. The Netherlands also lists unspecified PFC mix emissions from category 2B9. If we assume a similar mix as for Japan, this would contribute less than 0.6 t yr⁻¹ of *c*-C₄F₈.

Two countries report emissions of an unspecified mix of HFCs and PFCs and other fluorinated compounds to the UNFCCC, the United States of America (293 (1990) – 9449 (2014) Gg CO₂-eq.) and Germany (152 (2014) – 5773 (1995) Gg CO₂-eq.). The German NIR details that their emissions of unspecified mix of PFCs and HFCs and other fluorinated compounds is comprised of various HFCs, hydrofluoroethers (HFE), C₃F₈, higher PFCs, perfluorinated polyether (PFPE), anesthetics, and SF₆ from categories 2B9 and 2H3 (Others), but not *c*-C₄F₈.

The US reports *c*-C₄F₈ emissions of a few t yr⁻¹ from category 2E1 (IC or SC production). The NIR details that the US emissions of unspecified mix of PFCs and HFCs and other fluorinated compounds stems from category 2F6 (product uses as substitutes of ozone depleting substances (ODSs), other applications). From the description of category 2F and subcategory 2F6 it seems likely that *c*-C₄F₈ is at most a minor component of category 2F6 which is comprised of various HFCs, HFOs, C₄F₁₀, and a diverse collection of PFCs and PFPEs employed for solvent applications. Based on this one may conclude that no additional *c*-C₄F₈ emissions occur. However, data from the US EPA (https://www.epa.gov/sites/production/files/2018-10/ghgrp_i_freq_request_data_8_19_2018.xlsx, accessed Jan 2019) details that *c*-C₄F₈ emissions from three fluorochemical production facilities in the eastern US ranged from 29 to 62 t yr⁻¹ from 2011 to 2017. At least two of these facilities are known to produce TFE, HFP, and/or PTFE and it is likely that these facilities use the process via pyrolysis of HCFC-22, with *c*-C₄F₈ as an intermediate/by-product, which probably is the source of these reported emissions. These *c*-C₄F₈ emissions, which are ~8 times larger than the emissions listed *c*-C₄F₈ emissions from category 2E, are currently not reported in category 2B9 (fluorochemical production). The US EPA intends to report these emissions once emissions for the years 1990 to 2010 have been estimated (currently only data from 2011 onward exists) to fulfil UNFCCC reporting requirements to estimate emissions for each year since 1990 (US EPA, personal communication). It is unclear if these *c*-C₄F₈ emissions are currently reported in the unspecified mix of PFCs and HFCs and other fluorinated compounds.

In summary, data submitted to UNFCCC probably represent 10–30 t yr⁻¹ (0.01–0.03 Gg yr⁻¹) of *c*-C₄F₈ emissions, with 25–30 t yr⁻¹ (0.025–0.030 Gg yr⁻¹) from 2011 to 2014. After adding the US emissions from fluorochemical production listed by the US EPA, this increases substantially to 50–83 t yr⁻¹ (0.05–0.083 Gg yr⁻¹) from 2011 to 2014. It seems that large uncertainties remain due to difficulties disentangling the emissions of unspecified mixes of PFCs and mixes of HFCs/PFCs/other fluorinated compounds and perhaps unquantified or unaccounted for emissions.

The Emissions Database for Global Atmospheric Research (EDGAR) aims to estimate global emissions, including from countries not reporting to the UNFCCC, most notably China, South Korea, and Taiwan which may have significant *c*-C₄F₈ emissions from their electronics and PTFE industries. EDGAR v4.2 (EDGAR, 2010) estimates

global *c*-C₄F₈ emission from three sources (SC production, solvent use, fire extinguisher use), but only until 2010. From 1970 to 1985, EDGAR reports no *c*-C₄F₈ emissions, followed by a rise to a few t yr⁻¹ in the early 1990s and to ~25 t yr⁻¹ (~0.025 Gg yr⁻¹) in 2008, followed by a decline to ~20 t yr⁻¹ (0.02 Gg yr⁻¹) in 2010.

For Japan, *c*-C₄F₈ emissions reported by EDGAR are broadly consistent with those calculated from the UNFCCC NIR (see above) from the electronics industry alone (category 2E, assuming an increasing fraction of abatement from 2005 forward); therefore the potential emissions from category 2B9 (fugitive emissions from fluorochemical production) estimated above do not seem to be included in EDGAR.

For South Korea a NIR with data until 2013 can also be obtained (<http://www.gir.go.kr/eng/>). EDGAR *c*-C₄F₈ emission estimates are broadly consistent with those estimated from the South Korean NIR using IPCC methodologies for the electronics industries (category 2E), a few t yr⁻¹ until 2010.

For Taiwan we received *c*-C₄F₈ emissions from their NIR (Chang-Feng Ou-Yang, personal communications). Emissions reported by EDGAR are consistently lower than given in the NIR, at most a t yr⁻¹ versus a few t yr⁻¹ (since 2001) to ~20 t yr⁻¹ (2014). The Taiwanese NIR only includes emissions from SC, IC, and memory production, potentially excluding other emission sources, such as from LCD/TFT production. For China, Malaysia, and Singapore, EDGAR lists only very small emissions of less than a t yr⁻¹. Particularly for China this seems unlikely due to its large electronics and PTFE industries, potential *c*-C₄F₈ sources.

The World Semiconductor Council (WSC) estimates PFC emissions from their member industries in China, Taiwan, Europe, Japan, South Korea, United States, which range from 14 t yr⁻¹ in 2012 to 24 t yr⁻¹ in 2016.

Based on the available information discussed above we constructed a bottom-up inventory. First, we added for each year and each country emissions reported to UNFCCC and the estimated portion from unspecified mix of PFCs. For 2011 to 2014 we also added the U.S. emissions from fluorocarbon production reported by US EPA. Then we calculated the maximum for each year and each country from these augmented UNFCCC data, US EPA, EDGAR, and estimates from the NIRs, as detailed above. Globally these add up to 10–30 t yr⁻¹ (0.01–0.03 Gg yr⁻¹) from 1990 to 1999, 30–40 t yr⁻¹ (0.03–0.04 Gg yr⁻¹) from 2000 to 2010, and 100–116 t yr⁻¹ (~0.1 Gg yr⁻¹) from 2011 to 2014 (with a substantial fraction due to the U.S. emissions from fluorocarbon production reported by US EPA). WSC emissions discussed above corresponds to ~16 % of these global emissions for the years 2012 to 2014.

Supplemental figures

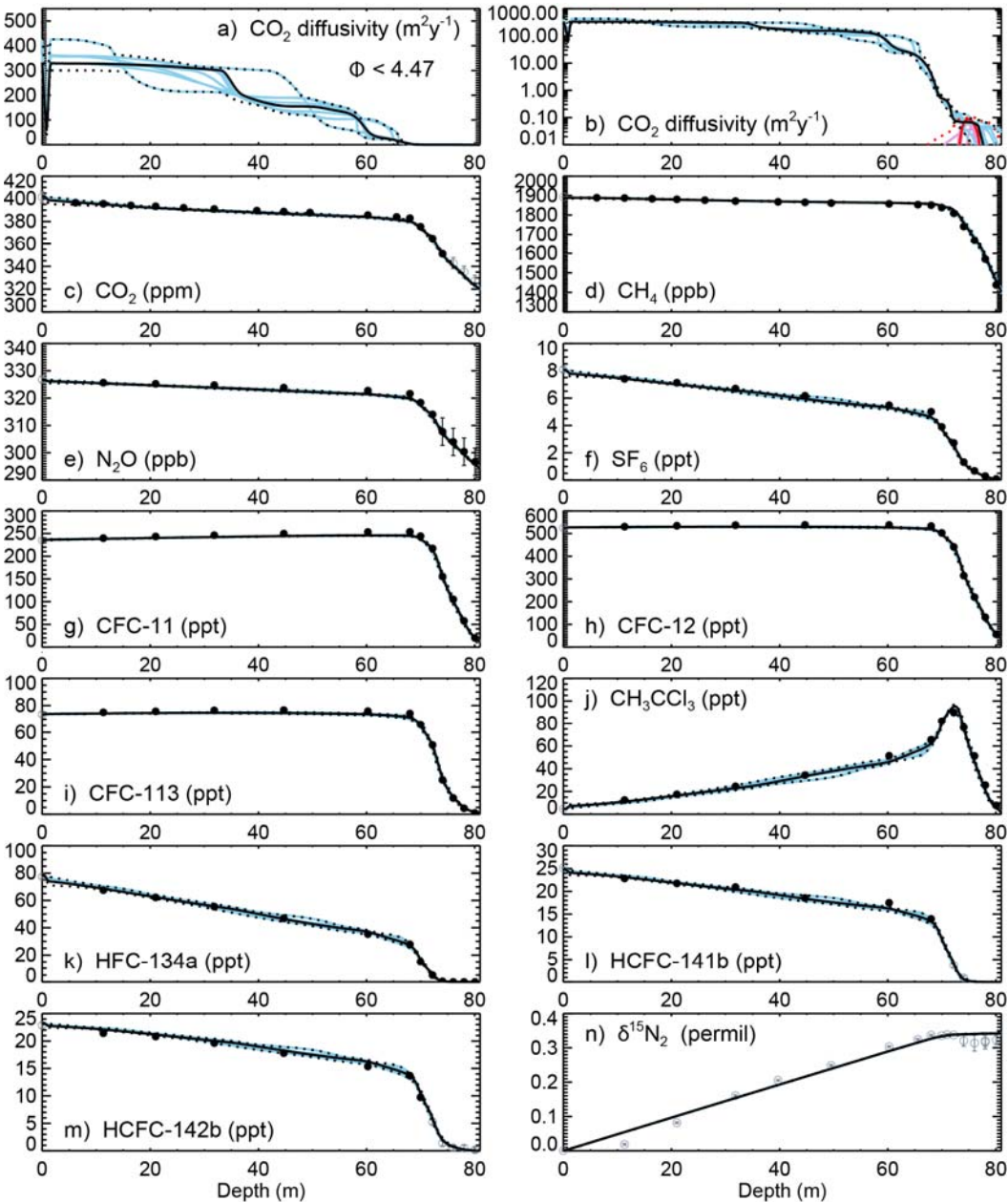


Figure S1. Tuning of the CSIRO firm model for the Summit13 site: CO₂ diffusivity on linear and log scales, and concentration profiles of CO₂, CH₄, N₂O, SF₆, CFC-11, CFC-12, CFC-113, CH₃CCl₃, HFC-134a, HCFC-141b, and HCFC-142b, as well as the δ¹⁵N₂ profile. The solid black lines show the case with the closest match to all observations used for diffusivity calibration. The dotted black lines show the upper and lower ranges of all cases that correspond approximately to a 68 % confidence interval. The blue curves show some representative cases within the 68 % confidence interval that are used in the CSIRO inversion to incorporate firm model uncertainty. In a) and b), the black and blue lines show molecular diffusivity, while in b), the red lines show dispersion in the lock-in zone (the red solid line is our best case, red dotted lines correspond to the 68 % confidence interval, and the pink lines show some representative cases). Measurements shown by black circles were used for calibration, and measurements shown by the grey circles were not used (see text).

We use firn air data for 11 tracers from the Summit13, Greenland site (CO_2 , CH_4 , N_2O , SF_6 , CFC-11, CFC-12, CFC-113, CH_3CCl_3 , HFC-134a, HCFC-141b, and HCFC-142b), to calibrate the diffusivity-depth profile and other diffusivity-related parameters in the CSIRO firn model using established methods. The firn model includes molecular diffusion throughout the firn (Schwander et al., 1993), and dispersion in the lock-in zone (Buizert and Severinghaus, 2016). The model gives the best match to observations with dispersion in the lock-in zone peaking at around $0.1 \text{ m}^2 \text{ yr}^{-1}$, consistent with Buizert et al. (2013) and Buizert and Severinghaus (2016), although there were also cases within the 68 % confidence interval that had no lock-in zone dispersion. We tested the use of eddy diffusion for convective mixing near the surface, but the best fit to observations was obtained without it, so it was not used in the final calibration. A melt layer was observed at Summit, due to melting that occurred in July 2012. The melt layer corresponds to a depth of around 60 cm but there were extensive percolated melt features down to around 1.5 m. The melt layer was included in the CSIRO firn model as described in Trudinger et al. (2013). Model layers in the CSIRO firn model move with the ice, and the timing of model layer generation at the surface was chosen so that the influence of the melt layer began in July 2012, and extended down to a model layer boundary that reached around 1.4 m at the time of firn sampling. The model produced the best fit to observations with reduction of diffusion by the melt layer of around 90 % (this value depends on the location of the model layers with time). We generated an ensemble of diffusivity parameters corresponding to a 68 % confidence interval as described in Trudinger et al. (2013), to use this to incorporate firn model uncertainty into the inversion.

Figure S1 shows the optimized diffusivity-depth profile and the modelled depth profiles for the calibration tracers. The atmospheric histories used to force the firn model were those compiled in Buizert et al. (2012) (based on Martinerie et al., 2009) for most tracers for calibration of firn models for the NEEM site, with extension to 2013 using in situ measurements from either Summit in the NOAA network (for CO_2 and CH_4) or Mace Head in the AGAGE network, with correction between the calibration scales used by NOAA and AGAGE where required. For N_2O , we used the NOAA record at Summit, extended prior to 1998 based on the Law Dome (SH) ice core record (Rubino et al., 2019) and the reconstruction by Prokopiou et al. (2017). For HCFC-141b and HCFC-142b, we compiled atmospheric histories using measurements from Mace Head from late 1994, and extrapolated back to zero before this. Due to the lack of information on atmospheric histories of these HCFCs before 1994, the deepest few measurements of these tracers were not used for calibration (indicated by the grey symbols in Figure S1). We used larger uncertainties for N_2O measurements prior to the in situ record due to the higher uncertainty in the atmospheric record. We excluded the deep CO_2 measurements from calibration, because at another NH site, NEEM, all models in the firn model intercomparison by Buizert et al., 2012 significantly underestimated CO_2 in the deep firn; the reason for this is not currently understood but could be due to in situ production or fractionation as discussed by Buizert et al. $\delta^{15}\text{N}_2$ is shown in Figure S1 but was not used for calibration because it is very sensitive to thermal effects at Summit that are not included in the firn model and not important for $c\text{-C}_4\text{F}_8$.

Buizert, C., Martinerie, P., Petrenko, V. V., Severinghaus, J. P., Trudinger, C. M., Witrant, E., Rosen, J. L., Orsi, A. J., Rubino, M., Etheridge, D. M., Steele, L. P., Hogan, C., Laube, J. C., Sturges, W. T., Levchenko, V. A., Smith, A. M., Levin, I., Conway, T. J., Dlugokencky, E. J., Lang, P. M., Kawamura, K., Jenk, T. M., White, J. W. C., Sowers,

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Rubino, M., Etheridge, D. M., Thornton, D. P., Howden, R., Allison, C. E., Francey, R. J., Langenfelds, R. L., Steele, P. L., Trudinger, C. M., Spencer, D. A., Curran, M. A. J., Van Ommen, T. D., and Smith, A. M.: Revised records of atmospheric trace gases CO₂, CH₄, N₂O and $\delta^{13}\text{C}\text{O}_2$ over the last 2000 years from Law Dome, Antarctica, *Earth Syst. Sci. Data Discuss.*, 2018, 1-30, 10.5194/essd-2018-146, 2018.

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Trudinger, C. M., Enting, I. G., Rayner, P. J., Etheridge, D. M., Buizert, C., Rubino, M., Krummel, P. B., and Blunier, T.: How well do different tracers constrain the firn diffusivity profile?, *Atmos. Chem. Phys.*, 13, 3, 1485-1510, 10.5194/acp-13-1485-2013, 2013.

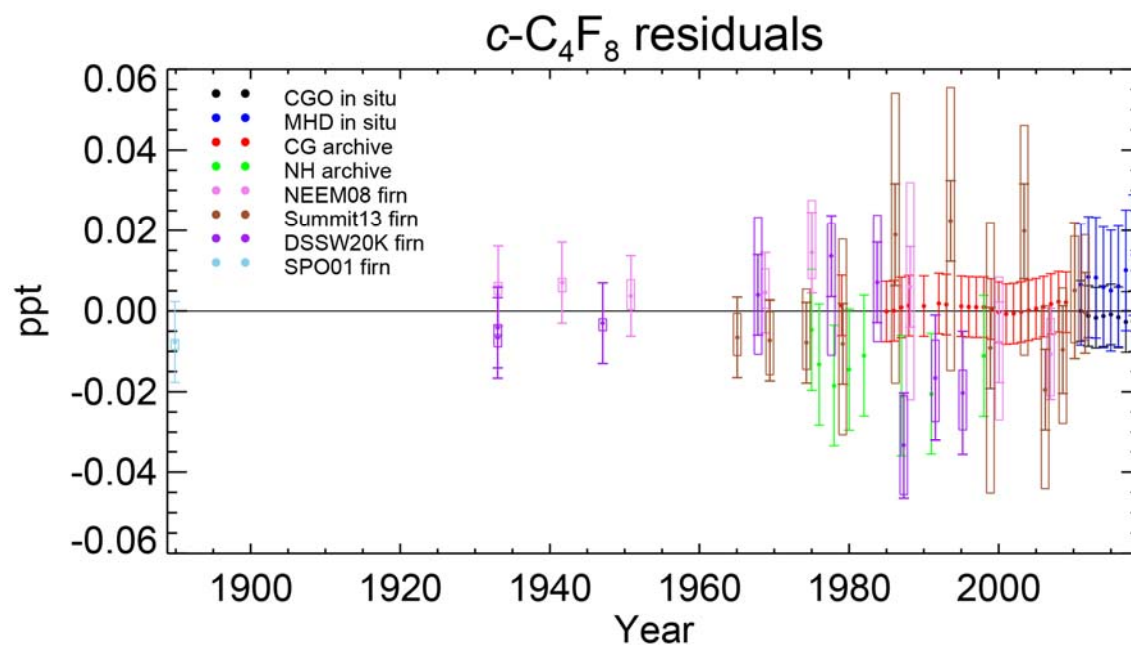


Figure S2. Residuals (model - observations) for the CSIRO inversion based on firn data and annual values from the smoothing spline in each hemisphere to in situ and archive data. The error bars show measurement errors used in the inversion (for the annual values this is the magnitude of correlated errors and for the firn data these are the measurement errors with a lower threshold of 0.01 ppt). The boxes show the range of uncertainties derived from the ensemble of Green's functions from the firn model.

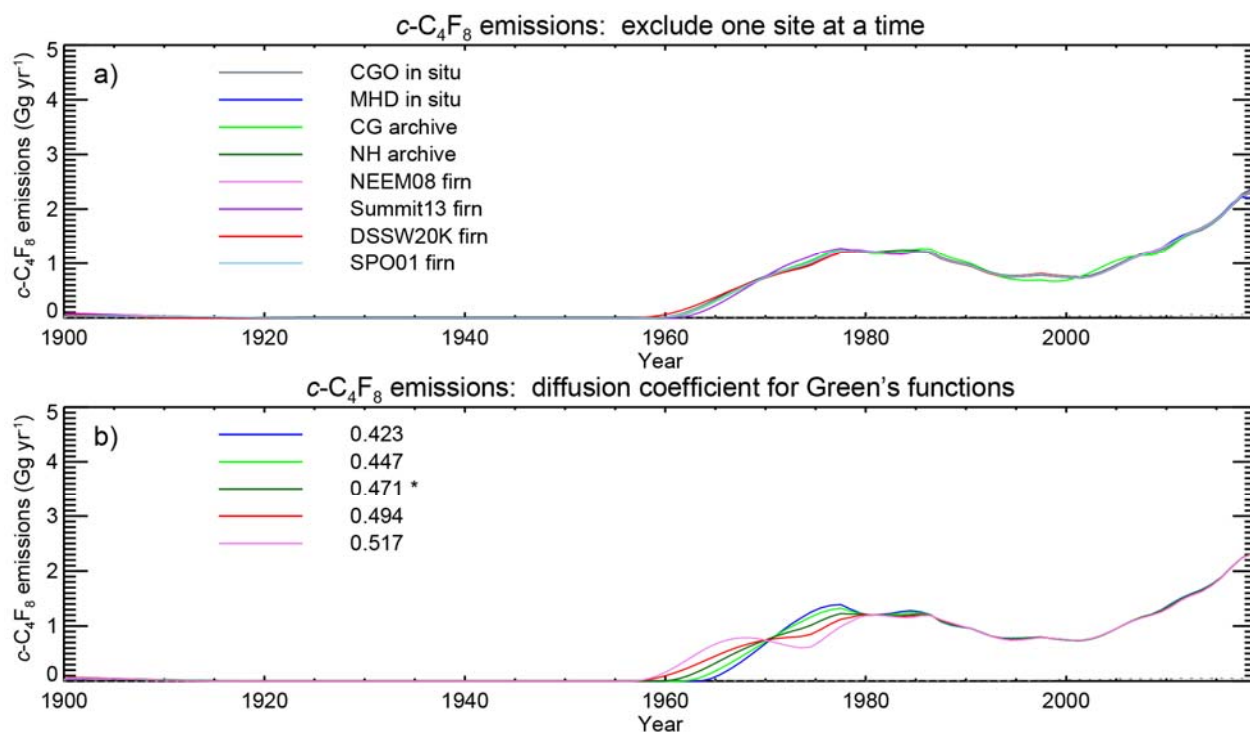


Figure S3. Sensitivity tests for the CSIRO inversion towards removal of individual data subsets and variation of the diffusion coefficient: a) inferred emissions with one firm site or in situ or archive part of the atmospheric record in each hemisphere left out at a time; b) inferred emissions for different values of the diffusion coefficient of $c\text{-C}_4\text{F}_8$ relative to CO_2 , with values from -10 % to +10 %.

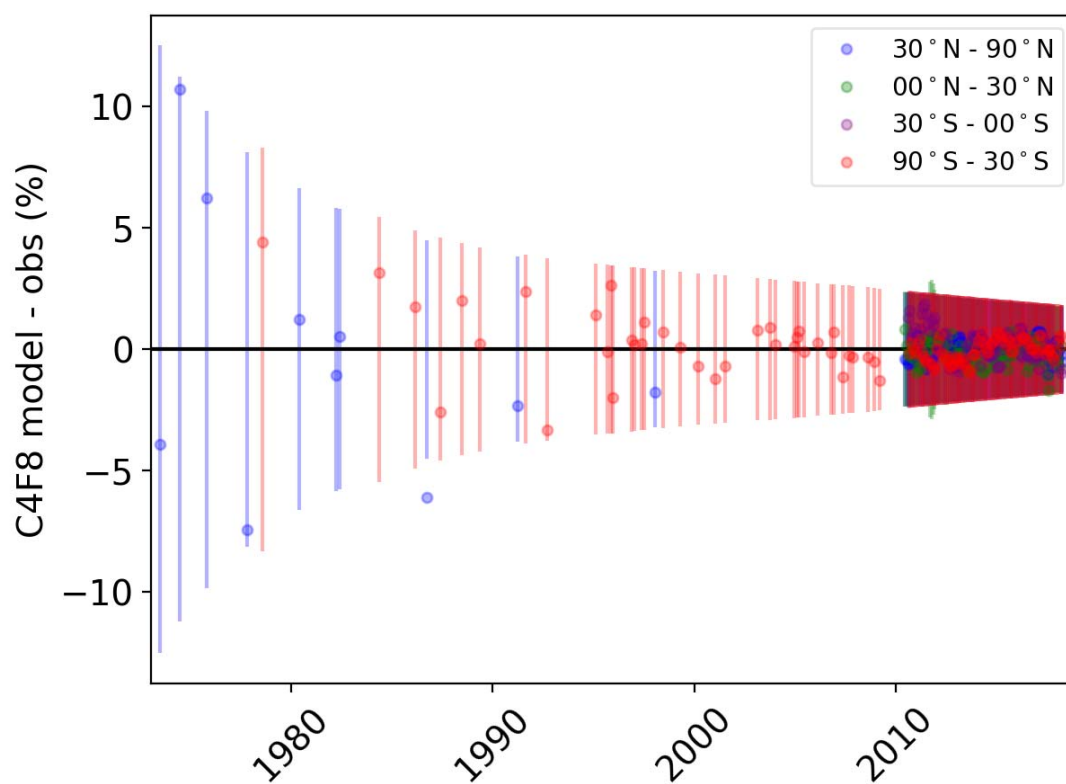


Figure S4. Residuals (model - observations) for the Bristol inversion based on archive and in situ data.

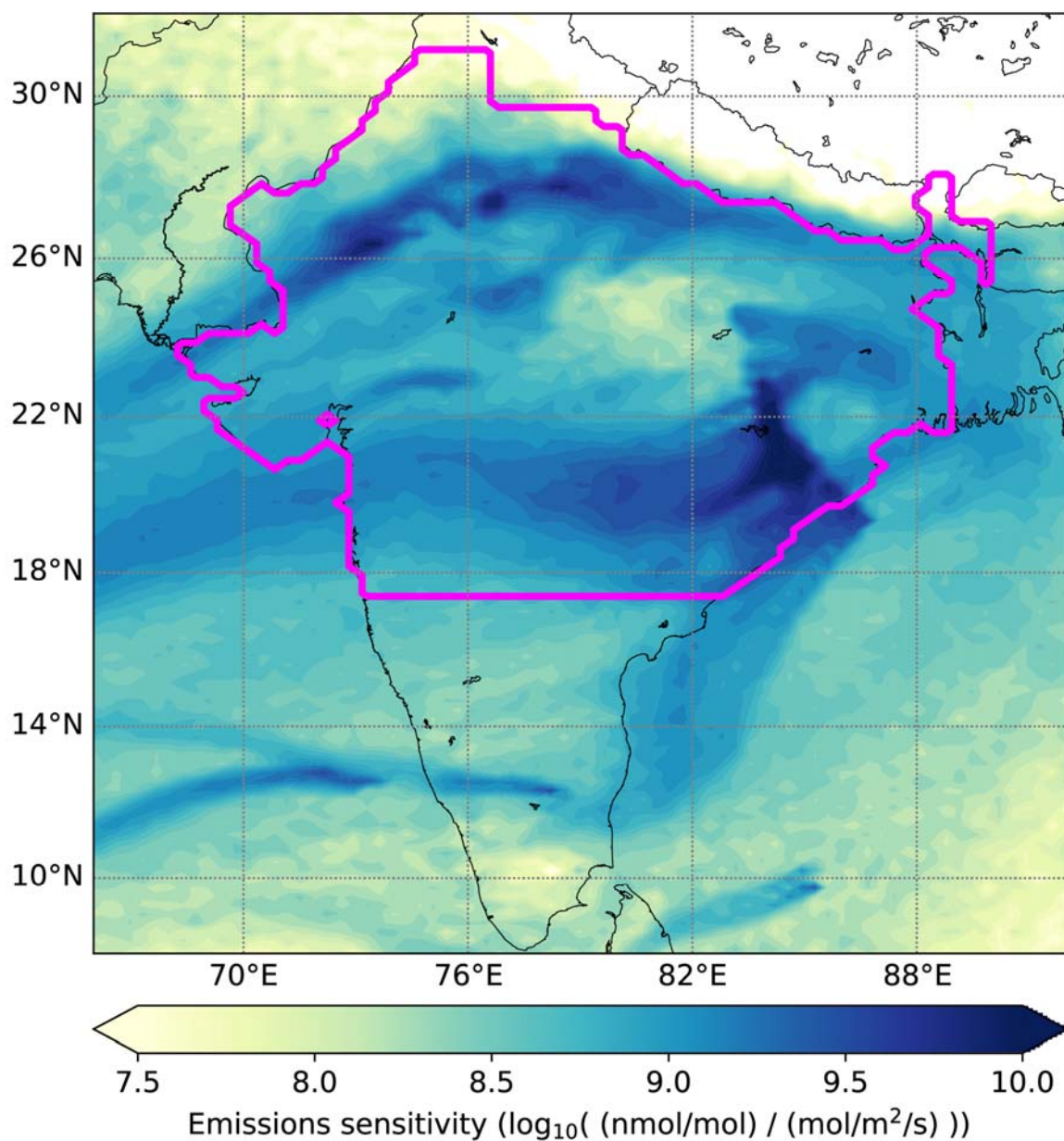


Figure S5: Emissions sensitivity averaged across all measurements made over the Indian subcontinent in June and July 2016. The region roughly corresponding to a maximum in emissions sensitivity is enclosed by the pink line. We denote this region as Northern and Central India (NCI).

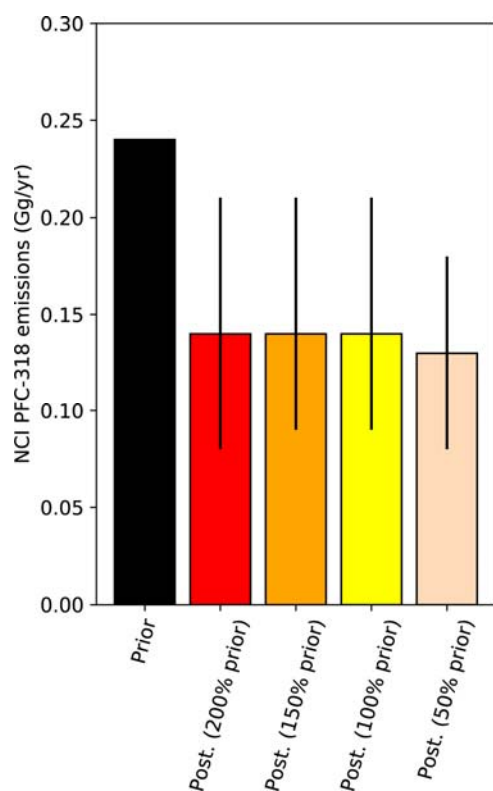


Figure S6: Sensitivity tests for the NAME-HB inversion for the Indian subcontinent. Emissions derived from priors of varying magnitudes (200% of the original prior, red bar, 150% of the original prior, orange bar, and 50% of the original prior, peach bar) indicate that $c\text{-C}_4\text{F}_8$ emissions determined for Northern and Central India are very insensitive to the choice of prior. The original prior (black bar) and posterior (yellow bar) estimates are also shown. For each estimate, error bars represent the 95% confidence interval of the posterior probability density function.

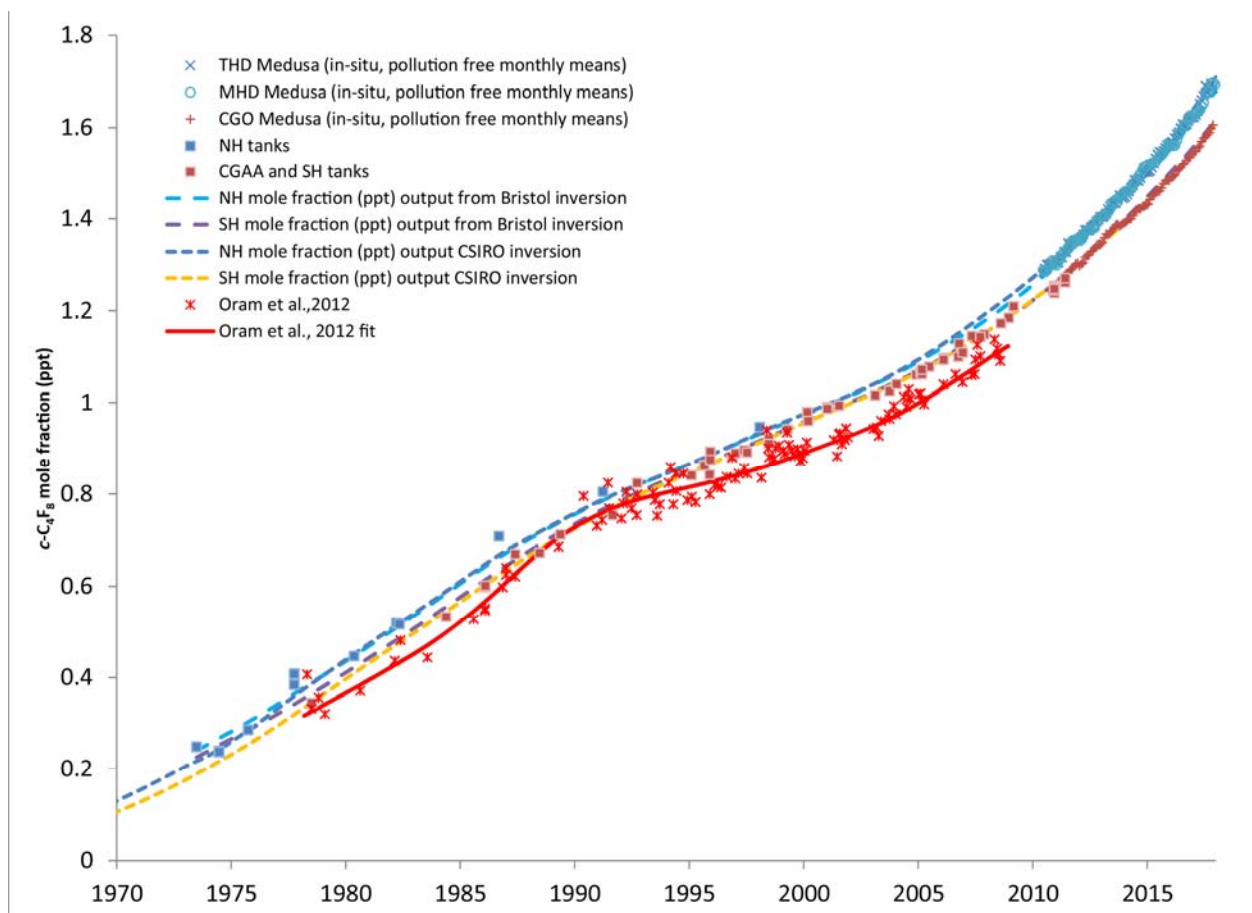


Figure S7. $c\text{-C}_4\text{F}_8$ mole fractions reconstructed here for the Northern (NH) and Southern Hemisphere (SH) compared to results from Oram et al., 2012 (SH only). Measured $c\text{-C}_4\text{F}_8$ mole fractions from THD, MHD, and CGO Medusa (in situ, pollution free monthly means, blue crosses and blue circles (NH), red pluses (SH) and NH (blue squares) and CGAA and SH tanks (red squares) are shown together with results from the Bristol (AGAGE 12-box, light blue (NH) and purple (SH) long dashes) and CSIRO (dark blue (NH) and orange (SH) short dashes) inversions. $c\text{-C}_4\text{F}_8$ mole fractions from Oram et al., 2012 (CGAA/SH only, red stars and red solid line) are shown for comparison.

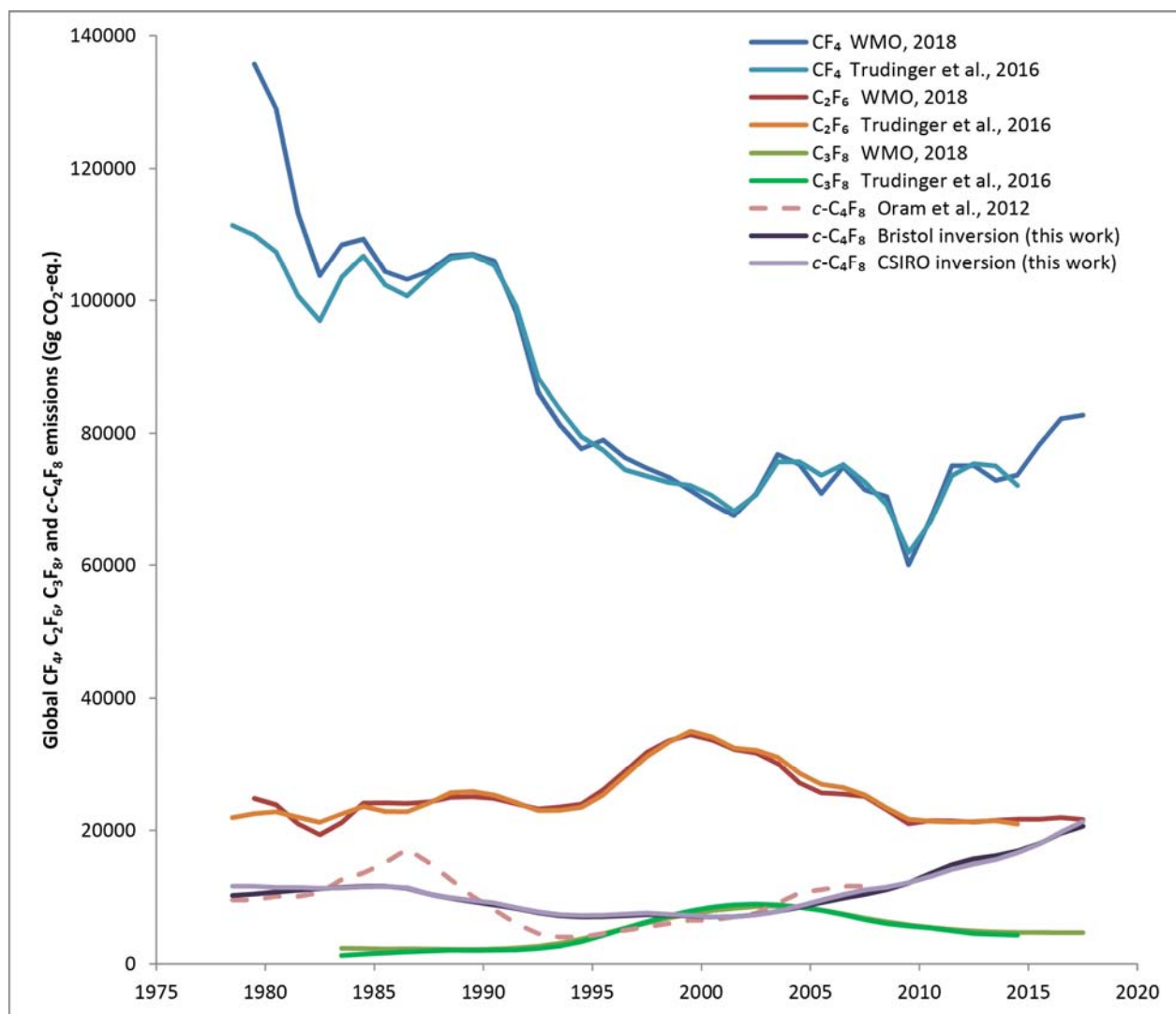


Figure S8: Global emissions of CF_4 , C_2F_6 , C_3F_8 , and $c\text{-C}_4\text{F}_8$ expressed as carbon dioxide equivalent emissions ($\text{CO}_2\text{-eq.}$) emissions (using GWP_{100}) (1.000.000 Gg $\text{CO}_2\text{-eq.}$ = 1 billion tonnes $\text{CO}_2\text{-eq.}$). In 2017, $c\text{-C}_4\text{F}_8$ emissions have reached 0.021 billion tonnes of $\text{CO}_2\text{-equivalent}$ compared to 0.083, 0.022, and 0.005 billion tonnes of $\text{CO}_2\text{-eq.}$ for CF_4 , C_2F_6 , and C_3F_8 .

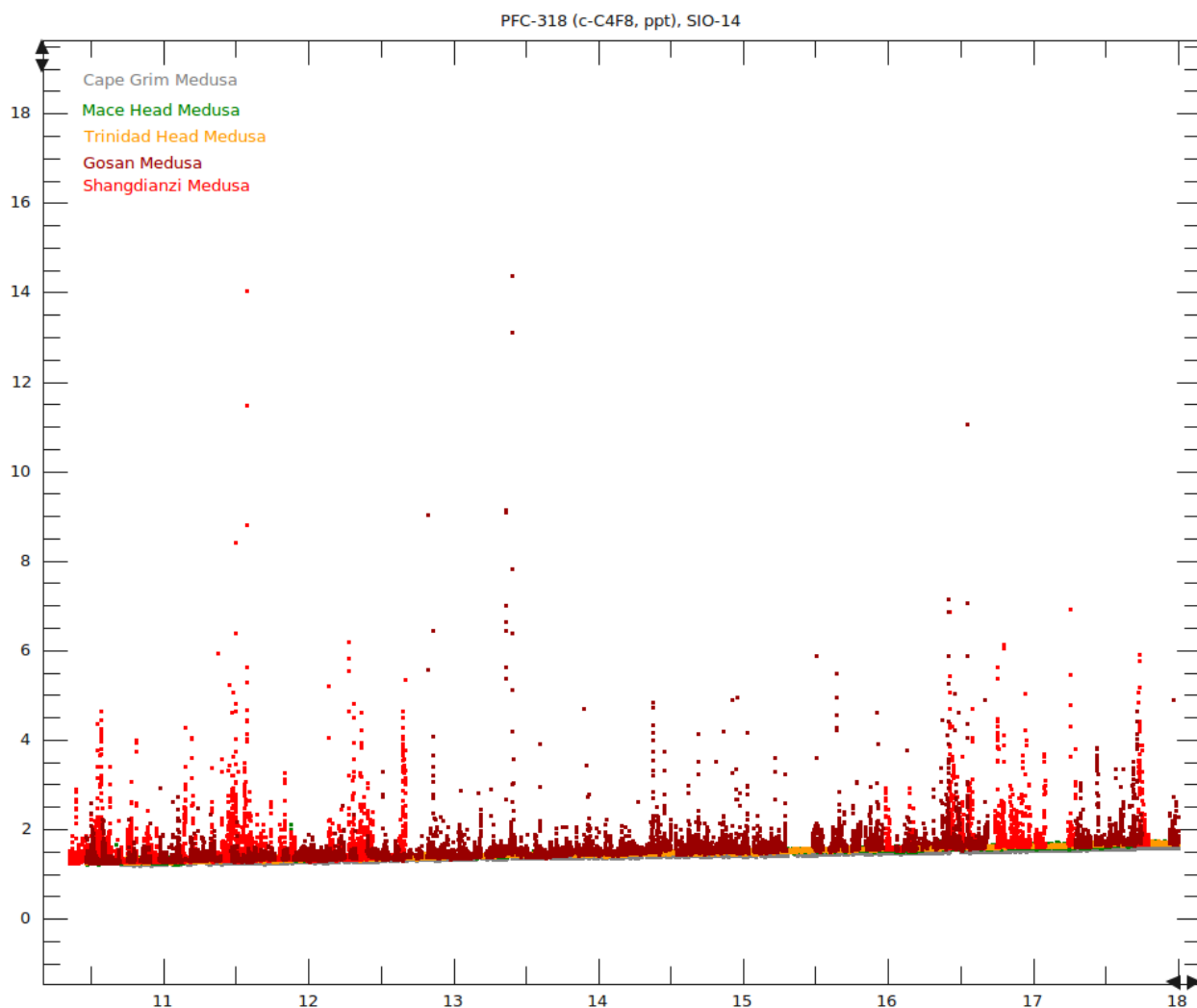


Figure S9. Magnitude of pollution events in East Asia. Among all stations of the AGAGE network, the two stations in eastern Asia, Gosan (brown) and Shangdianzi (red), show by far the most frequent and most pronounced pollution events of up to ~14 ppt above NH background (Mace Head, green, Trinidad Head, orange), indicating significant regional emissions. Measurements at Cape Grim, Australia (light grey), representing SH background, are also shown for comparison.

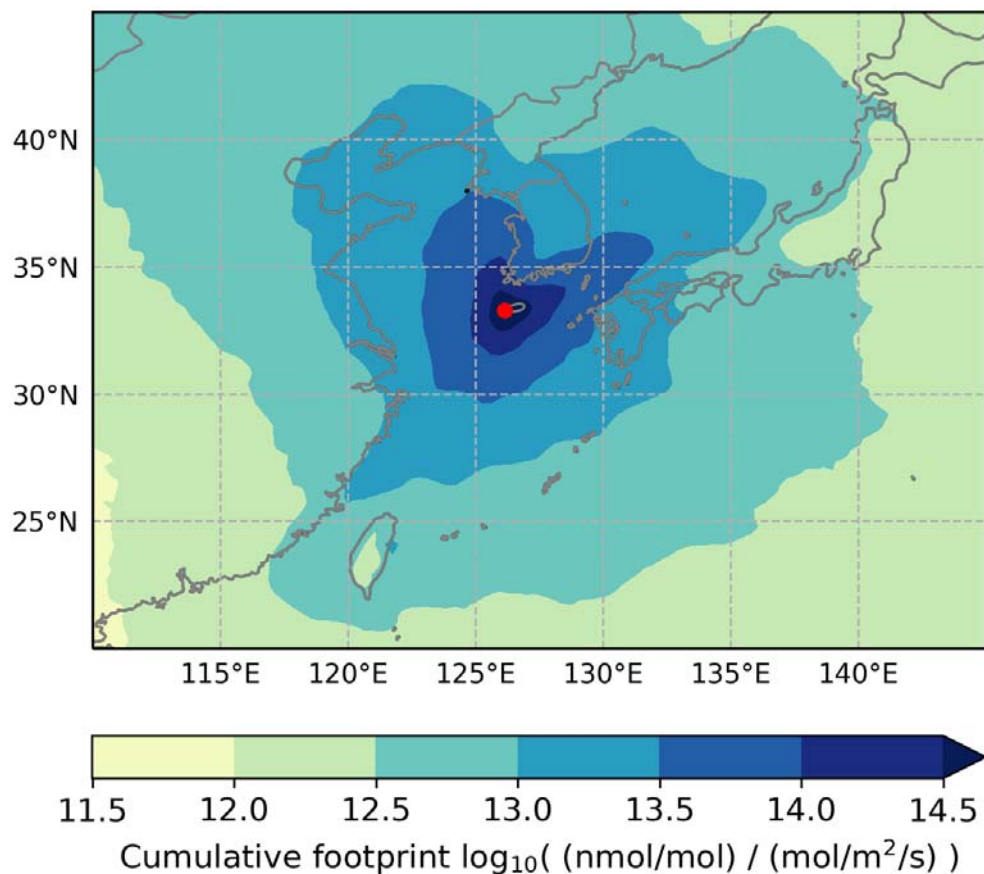
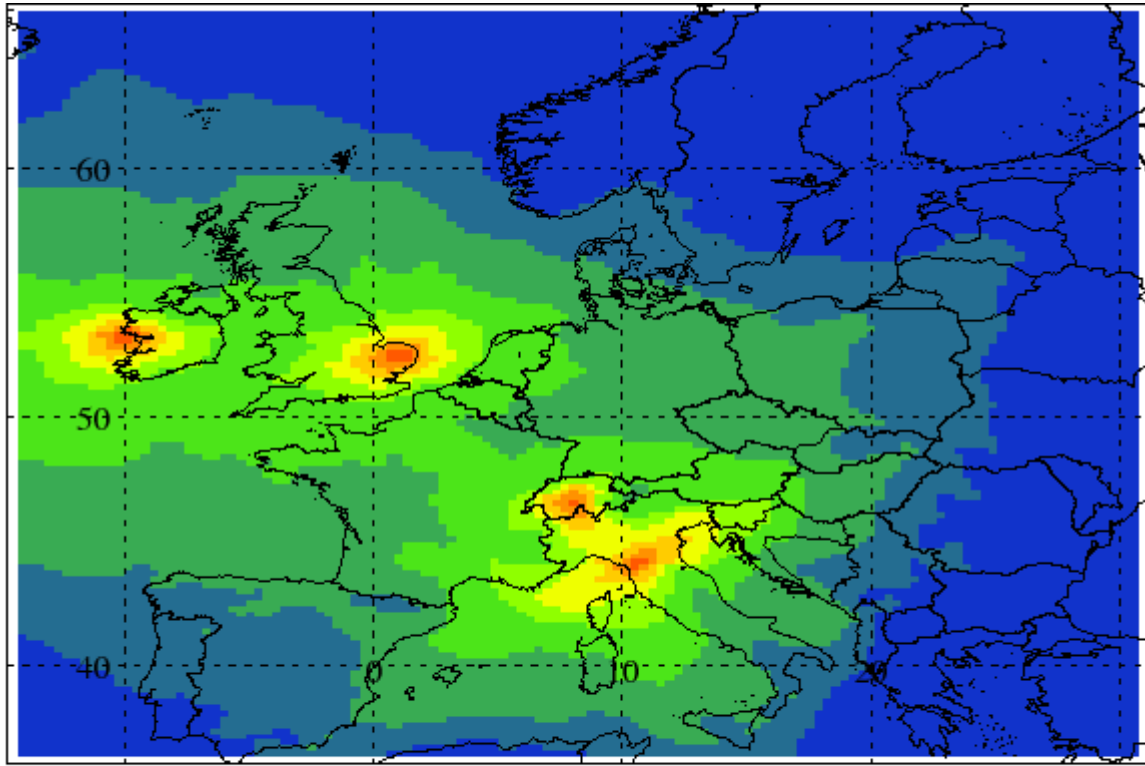


Figure S10: Cumulative footprint map for measurements at Gosan station, Jeju island, South Korea from 2010-2017 generated using the NAME transport model. The footprint indicates where the receptor station is sensitive to emissions. The sensitivity of the inversion generally decreases with distance to the receptor station resulting in relatively low sensitivity for emissions from western China, eastern Japan and Taiwan. Therefore, we report emissions for eastern China, western Japan, South Korea, North Korea, and parts of Taiwan. Eastern China contains the provinces Anhui, Beijing, Hebei, Henan, Jiangsu, Liaoning, Shandong, Shanghai, Shanxi, Tianjin and Zhejiang. Western Japan contains the prefectures Chugoku, Kansai, Shikoku and Okawa and Kyushu.



Maximum value = 4.93 g/m²/s



Figure S11: Cumulative footprint map for measurements at the Tacolneston, United Kingdom, Mace Head, Ireland, Jungfraujoch, Switzerland, and Monte Cimone, Italy stations for 2013. The sensitivity of the InTEM inversion is shown in arbitrary units. We report only estimated emissions from North Western Europe (42° N to 59° N and -11° E to 15° E) based on to the areas of highest sensitivity to the observations.

Footprint emission sensitivity on 19-Nov-2016 15:00:00

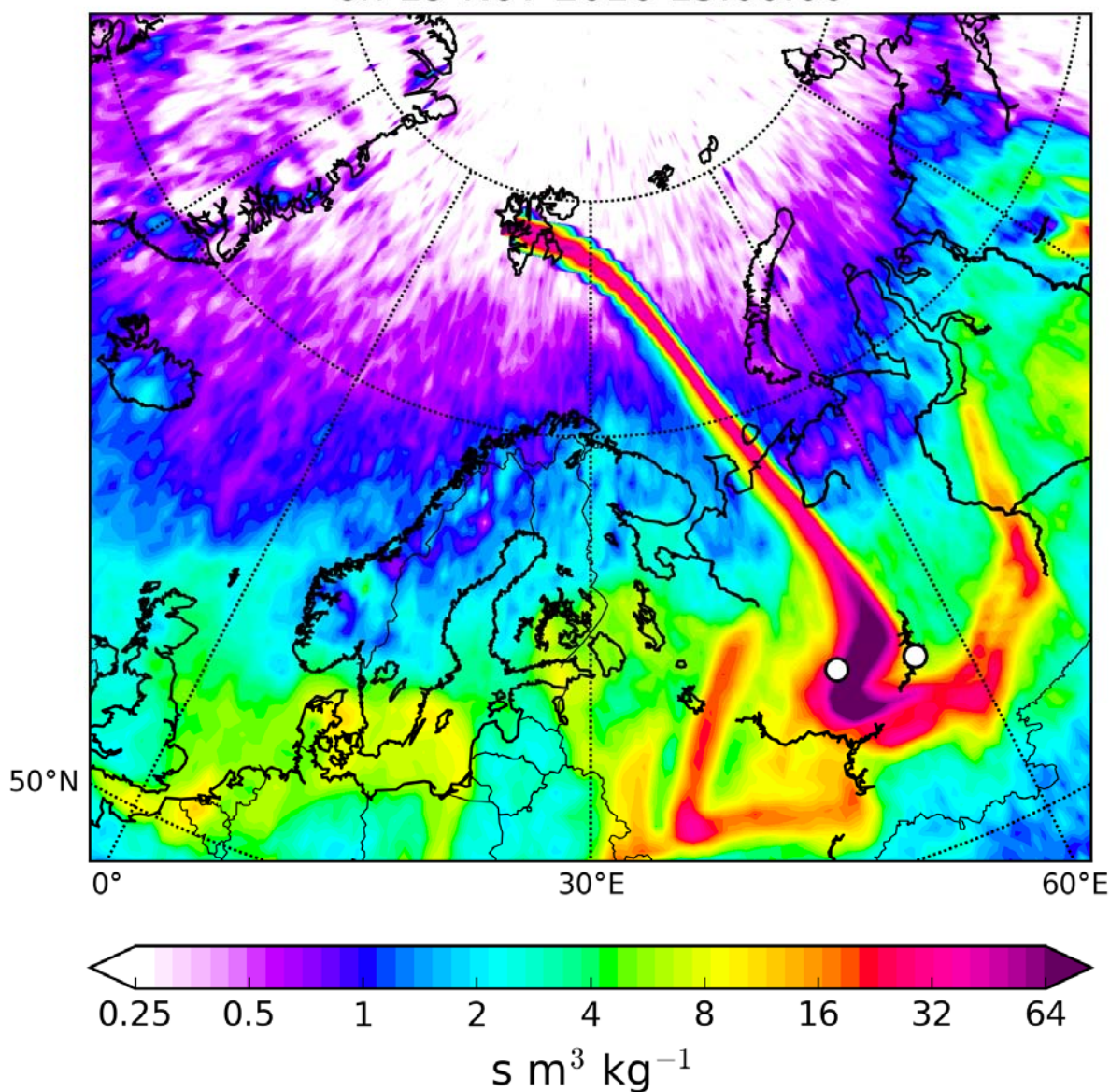


Figure S12. FLEXPART backward simulation for the strong $c\text{-C}_4\text{F}_8$ pollution event observed at Zeppelin station on November 19, 2016 indicate that this pollution event may be the result of direct transport of air from two facilities which produce PTFE and halogenated chemicals including $c\text{-C}_4\text{F}_8$ (HaloPolymer, Kirovo-Chepetsk, Kirov Oblast and Galogen Open Joint-Stock Company, Perm, Russia, each site marked with a white dot) to the Zeppelin station (marked with a white star).

Supplemental Tables

Table S1. The probability distributions assigned to the emissions and boundary conditions scaling and hyperparameters. Fixed parameters are those which have a fixed distribution that remain unchanged during the inversion. Hyperparameters represent the uncertainty in the uncertainties in the statistical model. These variable hyperparameters are estimated with their associated uncertainty within the NAME-HB inversion framework. This uncertainty translates into the total uncertainty in the posterior emissions estimates.

Parameter	Probability distribution	Fixed or variable?
Emissions and boundary conditions scaling	Log-normal(1,10)	Fixed
Model error (ppt)	Uniform(0.1, 10)	Variable
Correlation length scale (hours)	Uniform(1,120)	Variable
Number of Voronoi cells	Uniform(4,200)	Variable

Table S2. Chinese production of PTFE and FEP fluoropolymers (t yr^{-1}) and five year rise rates, including 2015–2020 forecast from the 13th Chinese five year plan

	2000	2005	2010	2011	2014	2015	2020*
PTFE	8,377	26,700	52,078	52,310	91,608	96,335	140,000
FEP			3,865		10,975	12,937	19,000
Fluoropolymers (total)			60,153		122,190	131,320	194,000
PTFE (% total)			87 %		75 %	73 %	72 %
FEP (% total)			6 %		9 %	10 %	10 %
% yr^{-1} increase		2000-2005	2005-2010			2010-2015	2015-2020*
PTFE		26 %	14 %			13 %	7.8 %
FEP						27 %	8.0 %

Source: www.qianzhan.com/analyst/detail/220/170629-c33a2ca7.html (Chinese, translate.google.com)

* Forecast from the 13th Chinese five year plan.

Table S3. Estimates of global PTFE market share by region

	2012 ⁺	2015 ⁺	2015 ⁺	2015 [#]	2015 [#]
North America	31 %	10 %		see RoW	
Europe	21 %	14 %		see RoW	
Asia Pacific (Total)	36 %	62 %		78 %	
China			53 %		67 %
Japan			9 %		11 %
Rest of World (Total)	12 %	14 %		22 %	
India			8 %		
Russia			6 %		
Total	100 %	100 %		100 %	

⁺www.industry-experts.com

[#]www.qianzhan.com/analyst/detail/220/170629-c33a2ca7.html

In 2015, PTFE production in China was estimated to account for 53 - 67% of global PTFE production.

Table S4. Estimates of global PTFE market share by company

	2012 ⁺		2015 [#]
DuPont	31.0 %	Dupont (Global incl. China)	13 %
Daikin	14.0 %	Daikin (Global incl. China)	13 %
Solvay	11.5 %	Solvay (China)	4 %
3M	9.0 %	3M	4 %
Others	31.0 %	Shangdong Dongyue Group	20 %
		Others (China)	30 %
		Others (Global excl. China)	11 %
Arkema SA	5.5 %		
Gujarat Fluorochemicals Ltd.	3.5 %		
		Asahi Glass (Japan)	5 %
Total	100.0 %		100 %

⁺www.industry-experts.com

[#]www.qianzhan.com/analyst/detail/220/170629-c33a2ca7.html

DuPont incl. Chemours, Solvay incl. Solexis, 3M incl. Dyneon, and Asahi Glass incl. AGC.