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Reconciling reported and unreported HFC emissions with atmospheric observations

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We infer global and regional emissions of five of the most abundant hydrofluorocarbons (HFCs) using atmospheric measurements from the Advanced Global Atmospheric Gases Experiment and the National Institute for Environmental Studies, Japan, networks. We find that the total CO_2-equivalent emissions of the five HFCs from countries that are required to provide detailed, annual reports to the United Nations Framework Convention on Climate Change (UNFCCC) increased from 198 (175–221) Tg-CO_2-eq·y^{-1} in 2007 to 275 (246–304) Tg-CO_2-eq·y^{-1} in 2012. These global warming potential-weighted aggregated emissions agree well with those reported to the UNFCCC throughout this period and indicate that the gap between reported emissions and global HFC emissions derived from atmospheric trends is almost entirely due to emissions from nonreporting countries. However, our measurement-based estimates of individual HFC species suggest that emissions, from reporting countries, of the most abundant HFC, HFC-134a, were only 79% (63–95%) of the UNFCCC inventory total, while other HFC emissions were significantly greater than the reported values. These results suggest that there are inaccuracies in the reporting methods for individual HFCs, which appear to cancel when aggregated together.

halocarbons | radiative forcing | climate change

Over the past two decades, emissions of hydrofluorocarbons (HFCs) have been growing rapidly as they replace the ozone-depleting chlorofluorocarbons and hydrochlorofluorocarbons (HCFCs). These compounds are used in applications such as refrigeration, air conditioning, and foam blowing, and as aerosol propellants and fire retardants (1). While the HFCs do not destroy stratospheric ozone, they are potent greenhouse gases, each with a considerable global warming potential (GWP) (2). As such, the HFCs were covered under the terms of the Kyoto Protocol to the United Nations Framework Convention on Climate Change (UNFCCC) (3), which places requirements on developed countries to provide detailed annual inventory reports of their emissions. The 42 countries that make up this group are referred to as Annex I countries.

The role of HFCs in contributing to global radiative forcing is estimated to be significant if emissions continue to rise as projected without any regulation (4). The inclusion of these non-ozone-depleting gases within the Montreal Protocol (5) has been proposed as a means of limiting their emissions via production/consumption controls, and therefore maintaining the climate benefits of the Protocol (6–8). The emissions reports that underpin the Montreal Protocol negotiations are calculated from a bottom-up perspective, where local activity data are combined with emission factors and aggregated to form the national inventory. These inventories are the subject of some uncertainty, resulting in calls for them to be verified by direct atmospheric measurements (e.g., refs. 9 and 10).

By examining global trends of HFCs in the atmosphere, it has been shown recently that aggregated global total HFC emissions were largely consistent with those reported by Annex I countries until the late 1990s (8). However, in recent years, inferred global HFC emissions have grown considerably faster than the Annex I reported estimates, to the extent that, in 2012, the total HFC emissions reported by the Annex I countries accounted for less than half of the global estimate inferred from observations (8). Despite efforts to use atmospheric measurements to derive emissions of individual HFCs on the global scale (e.g., refs. 11–14), or for certain regions (e.g., refs. 14–21), it is unclear to what extent this discrepancy in the aggregated total is due to the fact that many nations are not required to report HFC emissions, or because of errors in the reports themselves.

In this work, we seek to determine whether the reported emissions are accurate for five of the most abundant HFCs in the

Significance

Hydrofluorocarbons (HFCs) are among the atmosphere's fastest growing, and most potent, greenhouse gases. Proposals have been made to phase down their use over the coming decades. Such initiatives may largely be informed by existing emissions inventories, which, we show, are the subject of significant uncertainty. In this work, we use atmospheric models and measurements to examine the accuracy of these inventories for five major HFCs. We show that, when aggregated together, reported emissions of these HFCs from developed countries are consistent with the atmospheric measurements, and almost half of global emissions now originate from nonreporting countries. However, the agreement between our results and the inventory breaks down for individual HFC emissions, suggesting inaccuracies in the reporting methods for individual compounds.

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atmosphere: HFC-32 (CH_2F_2), HFC-125 (C_2HF_5), HFC-134a ($C_2H_2F_4$), HFC-143a ($C_2H_3F_3$), and HFC-152a ($C_2H_4F_2$). These HFCs are primarily used in air conditioning and refrigeration equipment, sources which, we assume, are relatively continuous and weighted toward population centers. We have not included HFC-23 in this analysis, because it is nearly exclusively emitted as a by-product of HCFC-22 production and therefore has very different emissions characteristics from the other major HFCs. Based on these individual HFC emissions estimates, we seek to determine the source of the discrepancy between estimated global HFC emissions and the total reported emissions. To determine the top-down emissions of these gases from reporting and nonreporting regions, a global trace gas monitoring network and 3D models of the transport and chemistry of these gases are required.

Using Atmospheric Measurements to Estimate Global and Regional HFC Emissions

To infer emissions, we used observations from two networks that measure all of the major HFCs at high frequency and high precision in the atmosphere: the Advanced Global Atmospheric Gases Experiment (AGAGE) (22) and the National Institute for Environmental Studies, Japan (NIES) (21). These networks are sensitive to many areas of the world for which HFC emissions are reported (shown by the blue areas in Fig. 1), and also to areas that remain unreported (yellow regions). We derived emissions from subregions within these two distinct areas using the observations and two chemical transport models.

Following ref. 23, the high-resolution, regional Numerical Atmospheric dispersion Modeling Environment (NAME) (24, 25) was used to simulate atmospheric HFC transport close to the monitoring sites. Simultaneously, the influence of changes to the global emissions field on all measurement stations was simulated using the global Model for OZone and Related Tracers (MOZART) (26) (*SI Text*). While the measurement network is relatively sparse in several regions of the world, the inversion system was designed to appropriately weight the emissions information that can be derived from global networks such as AGAGE and NIES. The high-resolution model was run within four regions, shown by red boxes in Fig. 1, outlining areas where the measurement stations were strongly influenced by regional HFC sources. These four areas were further divided into 155 subregions in total, from which emissions were derived. Outside of these four regions, sensitivities were calculated of modeled mole fractions to changes in continental emissions from seven "nonlocal" regions representing North America, South America, Europe, Africa, Oceania, Asian Annex I countries, and Asian non-Annex I countries. We estimated annual emissions for the period 2007–2012 from these 162 areas of the world and aggregated these emissions into two categories: those from Annex I countries and those from non-Annex I countries.

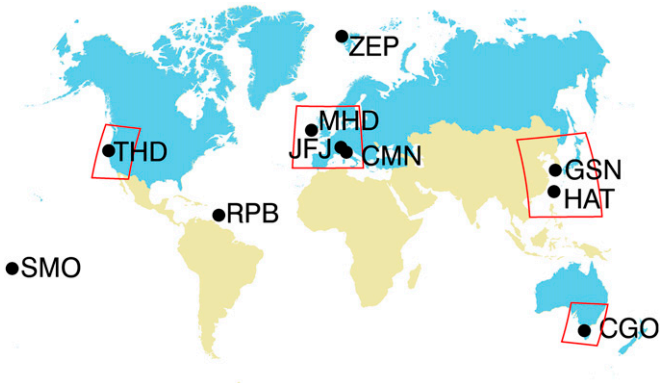

Fig. 1. Location of AGAGE and NIES stations, showing, from north to south: ZEP, Ny Ålesund, Norway; MHD, Mace Head, Ireland; JFJ, Jungfraujoch, Switzerland; CMN, Mt. Cimone, Italy; THD, Trinidad Head, United States; GSN, Gosan, South Korea; HAT, Hateruma, Japan; RPB, Ragged Point, Barbados; SMO, Cape Matatula, American Samoa; and CGO, Cape Grim, Australia. Red boxes indicate local regions where the NAME model was used with increased resolution compared with the global MOZART model, which was run for the remainder of the globe. Annex I countries are shaded blue, and non-Annex I are in pale yellow.

A hierarchical Bayesian framework was used to derive emissions and their uncertainties. This technique allows a more thorough exploration of uncertainties in the observations, prior constraints, and the chemical transport models than standard Bayesian methods (27) (see *SI Text*).

A priori emissions estimates were taken from the Emissions Database for Global Atmospheric Research (EDGAR v4.2) (28), which, like the UNFCCC reports, uses a bottom-up methodology, but shows different regional and temporal distributions. Recent work has revealed significant seasonality in global emissions of HFC-134a (29). We investigated whether our inversion of HFC-134a indicated a similar seasonality, and whether the derived annual emissions could be improved by accounting for seasonality in the prior emissions field. We further looked at the inclusion of seasonally varying prior emissions of HFC-125 and HFC-143a, as has previously been suggested (15). We did not investigate HFC-32 and HFC-152a as there was no previous suggestion of significant seasonality in emissions of these gases. We found closer agreement between the model and observations at northern hemisphere background sites for HFC-134a when summertime emissions were assumed twice as large as winter, as opposed to a constant annual field, in agreement with recent studies (29). However, when seasonal changes were included for HFC-143a, we found no such reduction in the model–measurement residuals, while the residual for HFC-125 increased slightly. In light of this, a seasonally varying prior emissions field was assumed for HFC-134a but a constant field was assumed in the inversion for HFC-125 and HFC-143a (see *SI Text* and Table S1). Since the focus of this work is on annual or multiannual scale emissions from large regions, our results were not appreciably affected by the inclusion, or otherwise, of this seasonality.

The top-down emissions estimates we present are the median of the posterior distributions obtained from our inversion with uncertainty ranges representing the fifth through 95th percentiles. Results presented in aggregate, in CO_2 equivalents, have been computed using the 100-y GWP (GWP_{100}) values from the Intergovernmental Panel on Climate Change, Second Assessment Report (30). Although these are not the most recent GWPs reported for the HFCs, these values are still used in the UNFCCC reports and were used throughout this work for consistency (see *SI Text* and Table S2).

Regional Attribution of Global HFC Emissions

Similarly to ref. 8, we find a dramatic rise in global emissions of the five HFCs during the study period (Fig. 2), from 303 (282–323) Tg CO_2-eq·y^{-1} in 2007 to 468 (436–500) Tg CO_2-eq·y^{-1} in 2012; a mean increase each year of 33 (22–44) Tg CO_2-eq (similar to the annual fossil fuel CO_2 emissions of New Zealand). As also shown in Fig. 2, our emissions estimates for only the Annex I countries agree with the reports to the UNFCCC remarkably well in terms of GWP_{100}-weighted emissions. The reports suggest that these emissions rose from 199 Tg CO_2-eq·y^{-1} in 2007 to 260 Tg CO_2-eq·y^{-1} in 2012, well within the uncertainties of our estimates of 198 (175–221) Tg CO_2-eq·y^{-1} in 2007 and 275 (246–304) Tg CO_2-eq·y^{-1} in 2012. This suggests that the UNFCCC reports provide an accurate representation of the Annex I HFC emissions when the five gases are aggregated together, and indicates that the previously noted discrepancy between global top-down and reported aggregate emissions is due primarily to the fact that many nations are not required to submit detailed annual emissions

reports. Indeed, we find that non-Annex I countries accounted for 42% (39–45%) of the total CO_2-equivalent emissions for these gases, averaged across 2010–2012. This is in contrast to the EDGAR estimates for 2007–2008, where non-Annex I aggregated emissions appear to be unrealistically small (Figs. 2 and 3).

Annex I Emissions: Right for the Wrong Reasons

In contrast to the high level of agreement of our estimates with the aggregated HFC reports, we derive significant discrepancies when emissions of these five species are analyzed individually (Fig. 3). Our estimated Annex I emissions of HFC-134a were found to be 21% (37–5%) lower than the UNFCCC inventory. In contrast, our estimates of the emissions of HFC-125 and HFC-143a were 20% (7–34%) and 33% (15–49%), respectively, higher than the inventory. There was no significant difference between the two Annex I estimates for HFC-32.

Estimated emissions of HFC-152a were more than 8 times greater than the inventory (Fig. 3). However, reporting of this compound is known to be incomplete, with several countries not reporting their emissions as a result of confidentiality considerations, due to the limited number of producers. Instead, emissions of HFC-152a from some countries, but not all, are included in a category of an "unspecified mix" of HFCs. We note that North America is the primary contributor to our derived Annex I total for HFC-152a, and that the absence of any such individual HFC-152a estimate submitted to the UNFCCC is likely to be the major reason for such a large discrepancy at the global scale (this region being responsible for approximately three quarters of global emissions in our inversion).

If we add this unspecified mix to the aggregate UNFCCC emissions of these five species, we obtain an 8–10% increase in the total reported (Fig. 2), reducing the agreement with our top-down aggregated emissions estimates by a similar margin (although still largely falling within our uncertainties). This would represent an upper limit of the error that these unspecified emissions would induce in the aggregated reports, as this category likely includes emissions of gases other than the five of interest here.

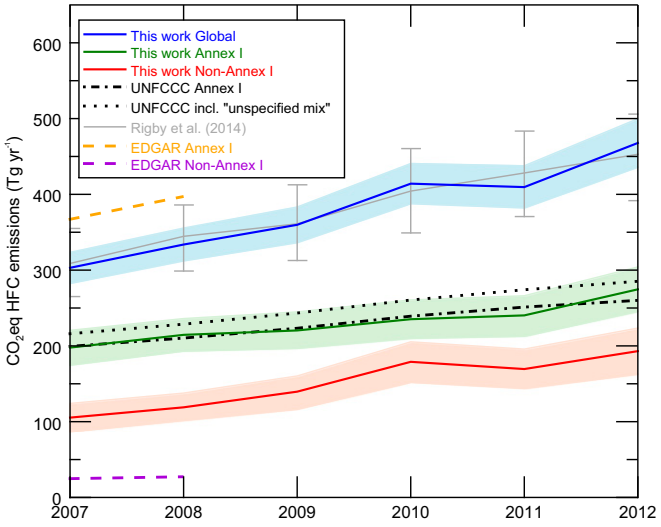

Fig. 2. Combined emissions of the five HFCs from 2007 to 2012 showing top-down estimates of global emissions (blue), Annex I emissions (green) and non-Annex I emissions (red). Shading reflects the fifth to 95th percentile of each estimate. UNFCCC emissions are shown as a black dash-dotted line, and UNFCCC reported emissions including an unspecified mix of HFCs are given by the black dotted line. EDGAR Annex I (orange dashed) and EDGAR non-Annex I (purple dashed) estimates are included for 2007–2008. Global estimates inferred from AGAGE data using a global 2D model from ref. 8 are also shown for comparison (gray).

Approximately 60% of emissions in the unspecified mix category were reported by Japan, almost all of which it attributes to the refrigeration and air conditioning sector. Since HFC-134a is the dominant source of emissions from this sector, it is highly likely that a disaggregation of the unspecified mix would result in reported emissions of HFC-134a being greater than the amount that is currently explicitly reported. If true, this would have the effect of increasing the apparent discrepancy with our estimates. Conversely, additional reporting of HFC-125 and HFC-143a in the unspecified mix category would reduce the disparity between the estimates of this compound. Therefore, it is likely that the incomplete reporting of individual HFC species by Annex I countries could explain some, but not all, of the discrepancy between our estimates and the reports of HFC-125, HFC-143a, and HFC-152a. However, it would increase the overreporting of HFC-134a and reduce the overall agreement of our estimates with the aggregate HFC emissions reports.

When combined into GWP_{100}-weighted CO_2 equivalents, the apparent overreporting of HFC-134a and the underreporting of HFC-125, HFC-143a, and HFC-152a cancel, such that the close agreement between reported aggregated HFC emissions and our top-down estimates is restored (notwithstanding the unspecified mix). However, when we analyze emissions from individual countries or subregions, we find that this pattern is not consistent across individual reporting nations (see *SI Text* and Table S3). Furthermore, the information provided to the UNFCCC shows that individual nations often assume significantly different emissions factors from each other, or use different activity metrics. Therefore, we propose that the close agreement in total Annex I aggregated HFC emissions is most likely to be primarily due to a fortuitous cancellation of errors in individual HFC reporting methods.

These results are shown by various tests to be largely independent of the a priori distribution of emissions, or the configuration of the measurement network (see *SI Text* and Figs. S1 and S2). Furthermore, inversions in which total UNFCCC emissions were assumed correct for each HFC provided significantly poorer agreement to the available measurements than our derived results (see *SI Text*, Table S4, and Figs. S3–S5). We note that the cancellation of errors applies only in terms GWP_{100}-weighted emissions and not in terms of total mass or moles of HFC emitted.

Emissions Trends from Annex I and Non-Annex I Countries

Table 1 shows our estimated Annex I emissions in gigagrams per year as well as the UNFCCC totals averaged across two time periods: 2007–2009 and 2010–2012. Annex I emissions of each HFC have increased across the two averaging periods. There is little or no trend in the differences between the UNFCCC inventory and our estimates for each gas, suggesting that the biases in the estimates discussed above have existed for the entire period investigated. The largest rises for Annex I countries, in percentage terms, were for HFC-32 and HFC-125, where increases of 63% (54–70%) and 44% (36–53%) were derived between these periods. This is likely to reflect the increased use of refrigerant blends containing these compounds, such as R410A (50% HFC-125, 50% HFC-32), in stationary air conditioning. The timing of this rise is in line with the phase-out of HCFC-22 use in new air conditioning units at the end of 2009, as mandated under the Montreal Protocol. Indeed, global growth of HCFC-22 emissions appears to have slowed since 2008 (8). R410A has been used as the primary replacement for HCFC-22 in stationary air conditioning, and this rapid rise in emissions of HFC-32 and HFC-125 might be expected to continue, as the phase-out of HCFC-22 is ongoing. In contrast to the rapid rise of these gases, emissions of HFC-143a and HFC-152a grew more moderately at 13% (5–21%) and 15% (12–18%), respectively. Smaller growth still was found for HFC-134a [9% (3–16%)], perhaps reflecting the influence of policy measures in Annex I countries such as the European F-gas and mobile air conditioning directives (31, 32).

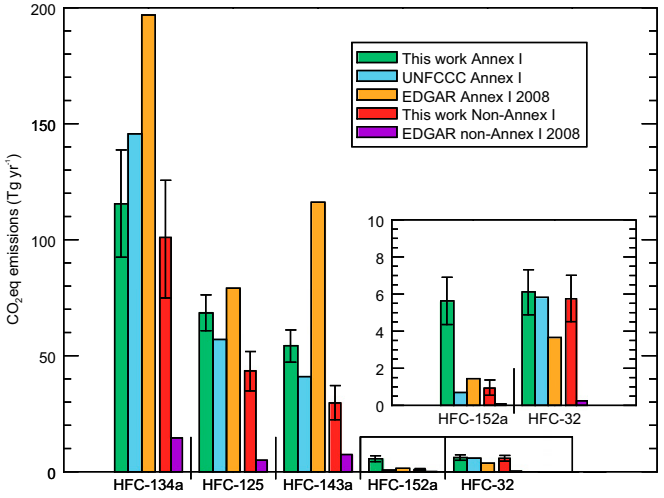

Fig. 3. Mean emissions during 2010–2012 of each HFC inferred for all Annex I countries (green) and non-Annex I countries (red). Also shown are the Annex I UNFCCC inventory (blue), EDGAR Annex I (orange), and EDGAR non-Annex I estimates (violet). EDGAR values are for 2008, the last year of available data. Uncertainty bars reflect the fifth to 95th percentile of the inferred emission estimates.

Table 1 and Fig. 2 also show the rapid growth of HFC emissions in non-Annex I countries. In a similar pattern to Annex I countries, the largest percentage increases were for HFC-32 and HFC-125, of 166% (149–189%) and 100% (77–132%), respectively, perhaps reflecting similar pressures to phase down HCFC-22 use. However, in contrast to the relatively slow growth of HFC-134a in Annex I countries, non-Annex I emissions grew by 29% (16–41%) in our estimates, perhaps reflecting the increased use of mobile air conditioning in these countries.

A significant proportion of the non-Annex I emissions are attributed to East Asian countries (which include China and South Korea) surrounding two of the measurement sites. Aggregated HFC emissions from this non-Annex I East Asian region averaged 42 (36–50) Tg CO_2-eq·y^{-1} from 2007 to 2009, rising to 60 (53–70) Tg CO_2-eq·y^{-1} from 2010 to 2012. These emissions comprised 33% (29–39%) of the non-Annex I total in the latter period and 14% (12–16%) of the global total. The size and growth of these estimates highlight that there was a significant contribution from East Asia emissions to the non-Annex I total, the monitoring of which could be important for future policy decisions. However, the results also indicate there are other areas of the globe that are likely to have significant emissions (such as south Asia, Africa, and South America) but are not well monitored by the current AGAGE and NIES networks. A further breakdown of regional and national emissions estimates is given in Table S3.

Future of HFC Emissions Verification

Owing to the relatively sparse nature of the measurement network, this paper has largely focused on aggregated emissions from large regions of the world (primarily total Annex I and non-Annex I emissions). We find that, at these scales, our results are robust to a range of sensitivity tests (see *SI Text* and Figs. S1 and S2), to well within our derived uncertainties. The tests show that errors in the a priori uncertainty, or due to sparse spatial sampling, are not likely to strongly influence our conclusions. However, uncertainties in the chemical transport model may be important, and methods that can more fully incorporate such factors into inverse methods must be developed in future. Furthermore, in the future, if we are to better understand the discrepancies we see in the HFC emissions reports, or monitor the effectiveness of various policy directives [e.g., the proposed changes to the Montreal Protocol, or the European mobile air conditioning directive (32)], it is important that we be able to derive emissions at national scales. As indicated in Fig. 1, there are several regions of the world to which the AGAGE and NIES networks are relatively insensitive at present (e.g., the east coast of the United States or South Asia). Therefore, in our analysis, uncertainties are relatively large for certain important regions. Recent studies have used expanded national networks to derive emissions of some of these species in certain regions [e.g., recently derived HFC-134a emissions from the United States using measurements from the National Oceanic and Atmospheric Administration (NOAA) network (33)]. If national emissions verification is to become more robust, such efforts will be needed across the globe.

Conclusions

We present global and regional emissions estimates of five of the major HFCs. Combined GWP_{100}-weighted emissions of these gases from Annex I countries were 250 (223–277) Tg CO_2-eq·y^{-1} averaged across 2010–2012 and were in good agreement with the UNFCCC inventory reports throughout the inversion period. This shows that the recently noted discrepancy between global emissions inferred from atmospheric measurements and the UNFCCC reported emissions is largely due to nonreporting countries. However, we find that this agreement in GWP_{100}-weighted aggregated Annex I emissions is due to a cancellation of apparent errors in the inventory estimates of individual HFCs rather than to species-specific accurate reporting. Our estimated Annex I emissions of HFC-134a during 2010–2012 were 21% (37–5%) lower than the UNFCCC inventory, while HFC-125 and HFC-143a emissions were, respectively, 20% (7–34%) and 33% (15–49%) higher than those reported. Incomplete reporting of some compounds owing to confidentiality considerations may

Table 1. Three-year averaged Annex I, UNFCCC, and non-Annex I emissions for each HFC in gigagrams per year

Compound	Years	This work Annex I	UNFCCC	Difference (UNFCCC minus this work)	This work non-Annex I
HFC-134a	2007–2009	82.0 (66.8–97.1)	108.0	26.0	59.9 (45.4–74.0)
	2010–2012	88.8 (71.1–106.7)	112.0	23.2	77.7 (57.6–96.6)
HFC-125	2007–2009	17.0 (14.9–19.0)	12.9	−4.0	7.8 (6.1–9.6)
	2010–2012	24.4 (21.7–27.2)	20.4	−4.1	15.5 (12.4–18.5)
HFC-143a	2007–2009	12.7 (11.0–14.2)	8.1	−4.6	4.9 (3.5–6.3)
	2010–2012	14.3 (12.4–16.1)	10.8	−3.5	7.8 (5.9–9.8)
HFC-32	2007–2009	5.9 (4.5–7.2)	4.5	−1.4	3.3 (2.3–4.2)
	2010–2012	9.4 (7.5–11.3)	9.0	−0.5	8.8 (6.9–10.8)
HFC-152a	2007–2009	35.2 (27.7–42.6)	4.5	−30.8	6.6 (4.3–9.2)
	2010–2012	40.2 (31.1–49.3)	5.0	−35.3	6.6 (3.9–9.8)

be partially responsible for some of this discrepancy. Emissions from Annex I countries accounted for 58% (55–61%) of the global total CO_2-equivalent emissions of the five HFCs averaged across 2010–2012. A range of sensitivity tests show that our results are robust at the global scales examined here. However, if we are to examine errors in emissions reports at the national level for all reporting countries, an expansion to the monitoring network will be required.

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1. Montzka SA, et al. (2011) Ozone depleting substances (ODS) and related chemicals. *Scientific Assessment of Ozone Depletion: 2010* (World Meteorol Org, Geneva), p 516.
2. Myhre G, et al. (2013) Anthropogenic and natural radiative forcing. *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, eds Stocker TF, et al. (Cambridge Univ Press, Cambridge, UK).
3. United Nations Framework Convention on Climate Change (1997) *Kyoto Protocol to the United Nations Framework Convention on Climate Change* (United Nations, Geneva).
4. Velders GJM, Fahey DW, Daniel JS, McFarland M, Andersen SO (2009) The large contribution of projected HFC emissions to future climate forcing. *Proc Natl Acad Sci USA* 106(27):10949–10954.
5. United Nations Environment Programme (1987) *The Montreal Protocol on Substances that Deplete the Ozone Layer* (United Nations Environ Prog, Nairobi, Kenya).
6. US Environmental Protection Agency (2013) *Benefits of Addressing HFCs Under the Montreal Protocol* (US Environ Prot Agency, Washington, DC).
7. Velders GJM, et al. (2012) Climate change. Preserving Montreal Protocol climate benefits by limiting HFCs. *Science* 335(6071):922–923.
8. Rigby M, et al. (2014) Recent and future trends in synthetic greenhouse gas radiative forcing. *Geophys Res Lett* 41(7):2623–2630.
9. Nisbet E, Weiss R (2010) Atmospheric science. Top-down versus bottom-up. *Science* 328(5983):1241–1243.
10. Weiss RF, Prinn RG (2011) Quantifying greenhouse-gas emissions from atmospheric measurements: A critical reality check for climate legislation. *Philos Trans R Soc A* 369(1943):1925–1942.
11. O'Doherty S, et al. (2014) Global emissions of HFC-143a (CH_3CF_3) and HFC-32 (CH_2F_2) from in situ and air archive atmospheric observations. *Atmos Chem Phys* 14(17): 9249–9258.
12. Vollmer MK, et al. (2011) Atmospheric histories and global emissions of the anthropogenic hydrofluorocarbons HFC-365mfc, HFC-245fa, HFC-227ea, and HFC-236fa. *J Geophys Res* 116(D8):D08304.
13. Miller BR, et al. (2010) HFC-23 (CHF_3) emission trend response to HCFC-22 ($CHClF_2$) production and recent HFC-23 emission abatement measures. *Atmos Chem Phys* 10(16):7875–7890.
14. Stohl A, et al. (2009) An analytical inversion method for determining regional and global emissions of greenhouse gases: Sensitivity studies and application to halocarbons. *Atmos Chem Phys* 9(5):1597–1620.
15. Miller JB, et al. (2012) Linking emissions of fossil fuel CO_2 and other anthropogenic trace gases using atmospheric ($^{14}CO_2$). *J Geophys Res* 117(D8):D08302.
16. Yao B, et al. (2012) In-situ measurements of atmospheric hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) at the Shangdianzi regional background station, China. *Atmos Chem Phys* 12(21):10181–10193.
17. Barletta B, et al. (2011) HFC-152a and HFC-134a emission estimates and characterization of CFCs, CFC replacements, and other halogenated solvents measured during the 2008 ARCTAS campaign (CARB phase) over the South Coast Air Basin of California. *Atmos Chem Phys* 11(6):2655–2669.
18. Li S, et al. (2011) Emissions of halogenated compounds in East Asia determined from measurements at Jeju Island, Korea. *Environ Sci Technol* 45(13):5668–5675.
19. Kim J, et al. (2010) Regional atmospheric emissions determined from measurements at Jeju Island, Korea: Halogenated compounds from China. *Geophys Res Lett* 37(12): L12801.
20. Stohl A, et al. (2010) Hydrochlorofluorocarbon and hydrofluorocarbon emissions in East Asia determined by inverse modeling. *Atmos Chem Phys* 10(8):3545–3560.
21. Yokouchi Y, et al. (2006) High frequency measurements of HFCs at a remote site in east Asia and their implications for Chinese emissions. *Geophys Res Lett* 33(21):L21814.
22. Prinn RG, et al. (2000) A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE. *J Geophys Res* 105(D14):17751–17792.
23. Rigby M, Manning AJ, Prinn RG (2011) Inversion of long-lived trace gas emissions using combined Eulerian and Lagrangian chemical transport models. *Atmos Chem Phys* 11(18):9887–9898.
24. Jones A, Thomson D, Hort M, Devenish B (2007) The UK Met Office's next-generation atmospheric dispersion model, NAME III. *Air Pollution Modeling and Its Applications* (Springer, New York), Vol 17, pp 580–589.
25. Manning AJ, O'Doherty S, Jones AR, Simmonds PG, Derwent RG (2011) Estimating UK methane and nitrous oxide emissions from 1990 to 2007 using an inversion modeling approach. *J Geophys Res* 116(D2):D02305.
26. Emmons LK, et al. (2010) Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4). *Geosci Model Dev* 3(1):43–67.
27. Ganesan AL, et al. (2014) Characterization of uncertainties in atmospheric trace gas inversions using hierarchical Bayesian methods. *Atmos Chem Phys* 14(8):3855–3864.
28. European Commission Joint Research Centre, Netherlands Environmental Assessment Agency (2011) *Emission Database for Global Atmospheric Research (EDGAR), Release Version 4.2*. Available at edgar.jrc.ec.europa.eu. Accessed June 6, 2014.
29. Xiang B, et al. (2014) Global emissions of refrigerants HCFC-22 and HFC-134a: unforeseen seasonal contributions. *Proc Natl Acad Sci USA* 111(49):17379–17384.
30. Albritton D, Derwent R, Isaksen I, Lal M, Wuebbles D (1995) Radiative forcing of climate change. *Climate Change 1995: The Science of Climate Change. Contribution of Working Group I to the Second Assessment Report of the Intergovernmental Panel on Climate Change* (Cambridge Univ Press, New York).
31. European Union (2006) Regulation (EC) No. 842/2006 of the European Parliament and of the Council of 17 May 2006 on certain fluorinated greenhouse gases (Text with EEA relevance). *Off J Eur Union L* 161:1–11.
32. European Union (2006) Directive 2006/40/EC of the European Parliament and of the Council of 17 May 2006 relating to emissions from air conditioning systems in motor vehicles and amending Council Directive 70/156/EEC (Text with EEA relevance). *Off J Eur Union L* 161:12–18.
33. Hu L, et al. (2015) U.S. emissions of HFC-134a derived for 2008-2012 from an extensive flask-air sampling network. *J Geophys Res* 120(2):801–825.