Chemosphere 283 (2021) 131208

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

A review of bottom-up and top-down emission estimates of hydrofluorocarbons (HFCs) in different parts of the world

Hannah Flerlage [a,*,1], Guus J.M. Velders [b,c], Jacob de Boer [a]

[a] Vrije Universiteit, Department of Environment and Health, Faculty of Sciences, De Boelelaan 1085, 1081 HV, Amsterdam, the Netherlands
[b] National Institute for Public Health and the Environment (RIVM), Bilthoven, the Netherlands
[c] Institute for Marine and Atmospheric Research Utrecht (IMAU), Utrecht University, the Netherlands

ARTICLE INFO

Handling Editor: Y Yeomin Yoon

Keywords:
Hydrofluorocarbons (HFCs)
Emission verifications
Bottom-up estimation
Top-down estimation
Global warming
Hydrofluoroolefines (HFOs)

ABSTRACT

Hydrofluorocarbons (HFCs) are widespread alternatives for the ozone-depleting substances chlorofluorocarbons and hydrochlorofluorocarbons. They are used mainly as refrigerants or as foam-blowing agents. HFCs do not deplete the ozone layer, but they are very potent greenhouse gases, already contributing to global warming. Since 2019 HFCs are regulated under the Kigali Amendment to the Montreal Protocol, which demands reliable emission estimates to monitor the phase-down. Quantification of emissions is performed with two methods: bottom-up from product inventories or data on chemical sales; or top-down, inferred from atmospheric measurements by inverse modelling or interspecies correlation. Here, we review and compare the two methods and give an overview of HFC emissions from different parts of the world. Emission estimates reported by the different methods vary considerably. HFC emissions of developed countries (Annex I) are reported to the United Nations Framework Convention on Climate Change. These bottom-up estimates add up to only half of global emissions estimated from atmospheric data. Several studies with regional top-down estimates have shown that this gap is not owed to large-scale underreporting of emissions from developed countries, but mostly due to emissions from developing countries (non-Annex I). China accounts for a large fraction of the emissions causing the gap, but not entirely. Bottom-up and top-down estimations of emissions from other developing countries that could identify other large emitters are largely unavailable. Especially South America, West-, Central- and East-Africa, India, the Arabian Peninsula and Northern Australia are not well covered by measurement stations that could provide atmospheric data for top-down estimates.

1. Introduction

Hydrofluorocarbons (HFCs) are widespread alternatives for the ozone-depleting substances (ODSs) chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs). They are used today in a variety of applications, mainly as refrigerants for cooling and air conditioning or as foam-blowing agents (Montzka et al., 2018). Other uses are in metered dose inhalers, aerosol spray cans, fire protection systems, and solvents (TEAP, 2019; Velders et al., 2012). ODSs are being phased out by the Montreal protocol, which is often described as a unique example of a global agreement that successfully averted an environmental crisis (Birmpili, 2018). The Montreal protocol was agreed upon in 1987 and is still the only UN treaty ratified by all 197 member states. The ozone layer is now on its way to recovery after countries made concerted efforts of control, regulation and substitution (Strahan and Douglass, 2018). Parties are required to report production, consumption, and trade data of the regulated chemicals to the United Nations Environment Program (UNEP) ozone secretariat on an annual basis (Ozone Secretariat, 2018).

As ozone-depleting substances are also very potent greenhouse gases, the protocol has been contributing to climate change mitigation as well. The climate benefit of the Montreal protocol was estimated to 10 Gt CO_2eq annual emissions avoided by 2010 (Velders et al., 2007). The corresponding radiative forcing avoided by regulating ODSs in the Montreal protocol amounts to about 35% of radiative forcing from CO_2 in 2010 (Velders et al., 2012).

Following the protocol, the phase-out of ozone-depleting CFCs was finalized in 2010 and the phase-out of the less, but still ozone-depleting

* Corresponding author.
E-mail address: h.flerlage@uva.nl (H. Flerlage).
[1] Present address: Institute for Biodiversity and Ecosystem Dynamics, University of Amsterdam, P.O. Box 94240, 1090 GE, Amsterdam, The Netherlands.

https://doi.org/10.1016/j.chemosphere.2021.131208
Received 19 March 2021; Received in revised form 7 June 2021; Accepted 10 June 2021
Available online 14 June 2021

HCFCs will be virtually completed in 2020 for non-Article 5 ("developed") countries and 2030 for Article 5 ("developing") countries, with only small amounts allowed afterwards for servicing (Ozone Secretariat, 2018). Article 5 countries under the Montreal protocol are those parties eligible to receive financial support from the Multilateral Fund, while non-Article 5 countries contribute to financial support, following the principles of responsibility and ability to act.

Atmospheric mole fractions of HFCs are growing rapidly, with an average rate of 1.6 ppt (parts per trillion) per year between 2012 and 2016 (Montzka et al., 2018). The annual growth rates between 2012 and 2016 are higher than between 2008 and 2012 for HFC-32, HFC-125, HFC-134a and HFC-143a, the most abundant HFCs in the atmosphere (Montzka et al., 2018; Yao et al., 2019). Factors contributing to this increase are the replacement of HCFC with HFCs following the HCFC phase-out under the Montreal Protocol, and a rapidly growing demand for automotive vehicles, air conditioners and refrigerators driven by economic growth, especially in Article 5 countries (Chaturvedi et al., 2015; Velders et al., 2015). An overview of common HFCs, their relevant properties and applications is given in Table 1.

HFC-23 is not considered here since it is emitted predominantly as a byproduct of HCFC-22 production. It is used intentionally only in a few specialized applications and the patterns of emission and necessary regulations are very distinct from the ones of other HFCs (Fang et al., 2016).

HFCs do not deplete the ozone layer like chlorine- or bromine-containing analogues do (Ravishankara et al., 1994). Recent findings show an indirect depletion potential due to radiative forcing increasing tropospheric and stratospheric temperatures, which alters atmospheric circulation and accelerates the catalytic ozone destruction cycle (Hurwitz et al., 2015). While this effect has limited impact, HFCs being halocarbons, are potent greenhouse gases (Ramanathan, 1975). Global warming potentials (GWPs) express the effect of a substance on global warming relative to CO_2, based on the mass of the substance emitted. HFCs have GWPs of up to several thousands and thus significantly contribute to global radiative forcing (Montzka et al., 2015). Millet et al. (2009) estimated that halocarbons make up 9% of US total CO_2eq GHG emissions and 32% of Mexican total CO_2eq GHG emissions. In 2016, the amount of HFC emissions globally was equal to about 1.5% of total emissions in CO_2-equivalents from all long-lived greenhouse gases such as CO_2 and N_2O, despite the comparatively low mole fractions of HFCs in the atmosphere (Montzka et al., 2018).

The C–F bonds in HFC molecules absorb in the atmospherically relevant region of the IR spectrum, causing the high contribution to radiative forcing of many HFCs in the atmosphere (Burkholder et al., 2020; Hodnebrog et al., 2013; Strong et al., 2018).

As a group of potent greenhouse gases, HFCs are subject to the Kyoto protocol and Annex I countries (countries committed to mitigate climate change, according to responsibility and ability to act; OECD and former Soviet countries) have to report annual emission data to the United Nations Framework Convention on Climate Change (UNFCCC). However, there are no specific regulations for the treatment of HFCs in the Kyoto Protocol. In 2016 the phase-down of production and consumption of HFCs was added to the Montreal protocol in the Kigali Amendment (Montzka et al., 2018). Under the Kyoto protocol, Annex I countries report emissions to the UNFCCC, parties to the Kigali Amendment of the Montreal protocol however, will report production and consumption data instead of emissions to UNEP (Ozone Secretariat, 2018). Most countries subject to Article 5 of the Montreal Protocol are non-Annex I countries in the framework of the Kyoto Protocol.

A baseline for the stepwise phase-down of HFCs is set for each party (country). The Article 5 parties are split into two groups. For Article 5 group 1 parties the baseline is defined as the HFC production/consumption averaged over the years 2020–2022 plus 65% of the HCFC consumption baseline. For Article 5 group 2, i.e., Bahrain, India, Iran, Iraq, Kuwait, Oman, Pakistan, Qatar, Saudi Arabia, and the United Arab Emirates, the baseline is defined as the average production/consumption in 2024–2026 plus 65% of the HCFC consumption baseline. For non-Article 5 countries the baseline is formed by the years 2011–2013 (+15% of the HCFC baseline) and a 10% reduction is set starting in 2019 (Ozone Secretariat, 2018). The non-Article 5 countries Belarus, the Russian Federation, Kazakhstan, Tajikistan, and Uzbekistan form an exception with +25% of the HCFC baseline instead of 15% and a 5% reduction in 2020 (Ozone Secretariat, 2018).

Without the Kigali amendment or any other kind of regulation of HFC production and use, severe impacts on the global climate were projected. Velders et al. (2015) projected that HFC production and consumption would have rapidly increased over the next decades, causing significant increases in HFC mole fractions in the atmosphere. The resulting radiative forcing from unregulated HFCs would have reached 0.22–0.25 W m^{-2} in 2050 according to a baseline scenario. Global warming of up to 0.3–0.5 °C by 2100 was foreseen to be caused by unregulated production and use of high GWP HFCs alone (Montzka et al., 2018; Xu et al., 2013). With the provisions of the Kigali amendment the contribution of HFCs to global warming is projected to less than 0.1 °C in 2100 (Montzka et al., 2018).

The success of the Kigali Amendment is important for protecting the

Table 1
Formulas, GWPs, lifetimes, atmospheric mole fractions, and main applications of common HFCs regulated under the Kigali amendment.

Name	Formula	GWP (100 year time horizon) (Montzka et al., 2018)	Lifetime (years) (Montzka et al., 2018)	Global annual mean atmospheric mole fraction in 2016 (ppt) (Montzka et al., 2018)	Main Applications
HFC-134a	CH_2FCF_3	1360	14	89.5 (89.3–92.1)	Refrigeration, AC, Mobile AC, Insulating foams (Velders et al., 2009), Aerosols (Brack, 2016)
HFC-32	CH_2F_2	705	5 .4	11.9 (11.2–12.6)	Refrigeration, AC (Velders et al., 2009)
HFC-125	CHF_2CF_3	3450	30	20.4 (20.1–20.8)	Refrigeration, AC (Velders et al., 2009) Fire suppression (Brack, 2016)
HFC-143a	CH_3CF_3	5080	51	19.2 (19.0–19.3)	Refrigeration, AC (Velders et al., 2009)
HFC-152a	CH_3CHF_2	148	1.6	6.67 (6.72-6.61)	Plastic foams, Aerosols (Velders et al., 2009)
HFC-227ea	CF_3CHFCF_3	3140	36	1.21 (1.17–1.24)	Extinguishing agent (Yao et al., 2019), Fire suppression (Brack, 2016)
HFC-236fa	$CF_3CH_2CF_3$	7680	213	0.15	Extinguishing agent (Yao et al., 2019)
HFC-365mfc	$CH_3CF_2CH_2CF_3$	810 (972)[a]	8.9 (10.9)[a]	0.94 (0.87–1.00)	Insulating foams (Velders et al., 2009)
HFC-245fa	$CHF_2CH_2CF_3$	880	7.9	2.43	Insulating foams (Velders et al., 2009)
HFC-43-10mee	$CF_3CHFCHF-CF_2CF_3$	1470	17	0.27	Solvent (Gobierno de México, 2018)

[a] Updated values according to Burkholder et al. (2020).

climate. Without global measures, it is expected that the consumption of HFCs will increase strongly within the next years, driven by a rising demand for air conditioning and refrigeration (Montzka et al., 2018). Hence, to limit their climate impact, it is important to control and regulate HFC use and to limit emissions to the atmosphere through a working global agreement.

Reliable emission estimates of HFCs are necessary to monitor the phase-down of HFCs under the Montreal protocol. Two approaches exist to estimating emissions: top-down, from atmospheric measurements, or bottom-up, from production and sales data. As Nisbet and Weiss (2010) point out, bottom-up emission estimates are generally more prone to errors and manipulation. Top-down estimates rely on meteorological data like wind speeds and directions to project atmospheric transportation and obtain emission values by inverse modelling (Nisbet and Weiss, 2010). The spatial resolution of emission estimates is relevant to be able to compare top-down emission estimates to bottom-up reports at a national scale. A country-level resolution of emission data furthermore enables the control of emission legislation and the evaluation of policy tools (Graziosi et al., 2017).

The 2018 scientific assessment of ozone depletion states that bottom-up emission estimates reported by the Annex I countries are accounting for less than half of top-down emission estimates on a global scale (Montzka et al., 2018). This gap in emissions could in principle be due to underreporting of Annex I countries, systematic differences in the estimation methods or large emissions from non-Annex I countries.

Under the Kigali Amendment, all parties to the Montreal Protocol must by 2020 report consumption and production of HFCs to UNEP (as of May 2021, 104 parties have reported HFC consumption for at least one year, data is published here: https://ozone.unep.org/countries/data-table). Thus, there is a growing number of studies on HFC use and emissions in different parts of the world, recently also many from East Asia. China and India are especially interesting in this context because of their large size and the growth potential of the air conditioning and refrigeration markets (Velders et al., 2009).

This review seeks to give an overview of the current state of information on HFC emissions in different parts of the world and compare the methods used to derive them.

In the following sections we present a description of top-down and bottom-up methods used to derive emission estimates and then compare the emission estimates resulting from the two different estimation approaches per HFC gas. Furthermore, we give an overview of HFC emissions and the status of information about them in different parts of the world. After providing a brief outlook on HFC alternatives, we conclude with key recommendations for future research. Collected HFC emission and consumption data are provided in the supplementary data file.

2. Top-down emission estimation

The top-down method estimates emissions from atmospheric measurement data. This approach is especially suitable for HFCs as they are entirely synthetic compounds. Thus, there is no background of naturally occurring fluxes, as it is the case for example for carbon dioxide, where large natural sinks and sources makes the estimation of anthropogenic emissions from atmospheric data more complex (Nisbet and Weiss, 2010). A general description of the top-down and bottom-up methods for the estimation of regional HFC emissions can be found in the supplementary information.

On a global scale, emissions can be estimated with simple box models or three-dimensional models, even from individual measurement sites (Stohl et al., 2010). Because of their sufficiently long lifetimes, HFCs are relatively homogenously distributed in the troposphere. For regional or country-level emission estimates on the other hand, a network of many atmospheric measurement stations is necessary, with different sensitivities to emissions from regional sources (Brunner et al., 2017).

Emission estimates on a regional scale considered here are derived by inverse modelling or interspecies correlation. Back trajectory methods have been employed to allocate emissions of various atmospheric pollutants, but inverse modelling has generally proven to yield more accurate results (Fang et al., 2018). Inverse modelling uses chemical transportation models and inversion algorithms to retrace emissions on a spatial grid (Stohl et al., 2009). Using interspecies correlation, ratios between measured HFC concentrations and concentrations of a substance of known emission flux, such as carbon monoxide, can also be employed to obtain emission estimates for defined geographical areas (Stohl et al., 2010).

2.1. Atmospheric measurements

The basis for both inverse modelling and interspecies correlation are atmospheric measurements. There are several networks of in-situ measurement stations around the world, for example the Advanced Global Atmospheric Gases Experiment (AGAGE; http://agage.mit.edu/; Prinn et al., 2000), and the network of the National Institute for Environmental Science (NIES; https://db.cger.nies.go.jp; Yokouchi et al., 2006) in Japan. An overview of measurement stations is presented in Fig. 1. All networks provide high frequency measurements of HFCs. To quantify HFCs in the atmosphere, automated low-temperature pre-concentration and re-focusing are employed before the gases are measured with automated GC-MS (Graziosi et al., 2017; Lunt et al., 2015; Miller et al., 2008; Stohl et al., 2009).

The US National Oceanic and Atmospheric Administration's (NOAA) Global Greenhouse Gas Reference Network maintains intensive wide-range air flask sampling across the USA and central analysis of the samples in one of two GC-MS instruments (NOAA HATS (Halocarbon and other Atmospheric Trace Species); https://www.esrl.noaa.gov/gmd/; Hu et al., 2015). Fortems-Cheiney et al. (2015) include a list of global atmospheric measurement stations that quantify HFC-134a. Next to the permanently installed stations, measurements are performed on air sampled on aircraft campaigns (Millet et al., 2009; Say et al., 2019; Wofsy, 2011; Xiang et al., 2014).

Atmospheric HFC concentrations can be subject to seasonal variability (Xiang et al., 2014). Kuyper et al. (2019) found this effect for HFC-152a at Cape Point and reasoned that the seasonal variability is likely due to the comparably short lifetime of HFC-152a and the minimum winter hydroxyl radical concentration in the troposphere.

2.2. Inverse modeling

Inverse modelling enables the estimation of emissions from atmospheric data based on source-receptor relationships. This approach uses Lagrangian Particle Dispersion models (LPDMs) to trace back the origin of increases in HFC mole fractions determined from atmospheric data from geographically distributed measurements (Lunt et al., 2015). To obtain regional fluxes, an inversion algorithm is fed with the output of the LPDM, measurement data from different sites and bottom-up emission data as a priori information and their respective uncertainties (Stohl et al., 2010). Scheme 1 illustrates the process of inverse modelling.

The time frame used for the backwards simulation of the LPDMs vary. Say et al. (2019) employed the NAME (Numerical Atmospheric dispersion Modeling Environment) model with a 30-day backwards modulation of surface fluxes, when estimating Indian HFC emissions. This is a 10-day longer backwards modeling time than employed by Stohl et al. (2010), who used FLEXPART (FLEXible PARTicle dispersion model) for their inversion model. Kuyper et al. (2019) used NAME to evaluate emissions from South Africa, also employing 30-day backwards trajectories.

Lunt et al. (2015) used the models NAME and MOZART (Model for Ozone and Related Tracers) to infer HFC emissions from Annex I and Non-Annex I countries. MOZART simulates the global changes in emissions, whereas the NAME model is providing a higher spatial resolution for the area closer to the measurement site.

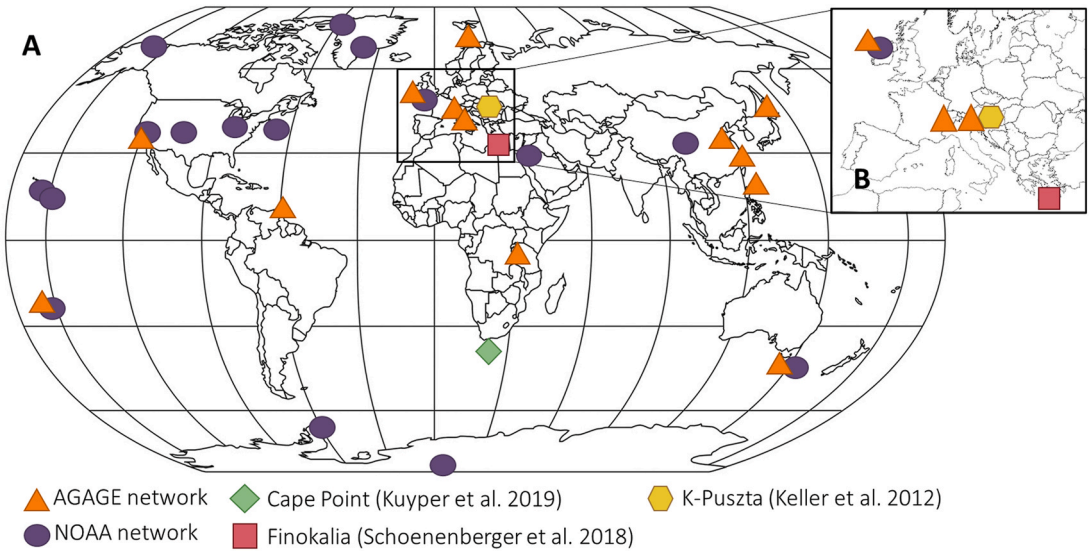
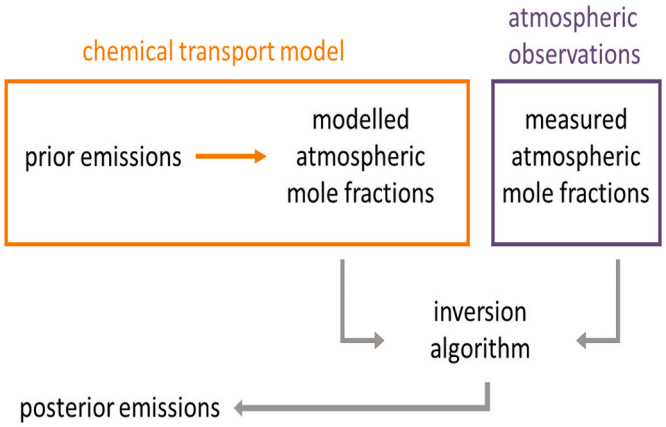


Fig. 1. A. Geographical distribution of sampling sites from the Advanced Global Atmospheric Gases Experiment (AGAGE), including stations from Japan's National Institute for Environmental Studies (NIES), and the US NOAA HATS flask sampling program (ESRL). Some further measurement sites have been used to measure HFCs as described in this review, in Cape Point, South Africa (Kuyper et al., 2019), K-Pustza, Hungary (Keller et al., 2012) and Finokalia, Greece (Schoenenberger et al., 2018). B. In Europe, the density of measuring stations is highest.



Scheme 1. Principle of top-down emission estimation using inverse modelling. A chemical transport model provides modelled atmospheric mole fractions from prior emission estimates which are fed to an inversion algorithm together with measured atmospheric mole fractions. The inversion algorithm matches the modelled atmospheric mole fractions to the measured ones by adjusting the emissions in the chemical transport model. So, the posterior emissions are generated as output of the inverse modelling.

To obtain prior data for Article 5 countries for their model, Schoenenberger et al. (2018) subtracted all reported emissions from Annex I countries to the UNFCCC from global emission estimates and then divided the rest to the non-Annex I countries according to population density. For Annex I countries, a priori emission estimates were taken from reports to the UNFCCC. In another study, no specific, spatially resolved emission inventory was available for China, hence global total emissions of HFCs in 2011 were used (Yao et al., 2019). These were likewise disaggregated resembling approximate population densities. A spatial distribution of emissions oriented at data of night light distribution from NOAA, serving as a proxy for population density, was used as prior in several studies (Kuyper et al., 2019; Say et al., 2019).

With the advance of analytic techniques, more detailed monitoring is becoming accessible, also at a city level (Ghandehari et al., 2017). This higher spatial resolution bears opportunities for better control and enforcement of regulations also on sub-national scales. Furthermore, it generally increases the traceability and understanding of emission sources and could activate sub-national regulatory actors.

2.3. Error sources and uncertainty of inverse modelling

Atmospheric measurements of HFC mole fractions are relatively precise with only small errors from the GC-MS analysis of the air samples in the order of about 2%. The determination of background level atmospheric mole fractions poses a relevant, but mostly not dominant source of uncertainty (Hu et al., 2017).

Inaccurate model simulations are causing relatively high uncertainties of emission values derived by inverse modelling (Yao et al., 2019). Multiple emission estimates from uncorrelated inversions were generated for example by Say et al. (2016) when estimating UK's HFC-134a emissions from atmospheric measurements and can be used to ascribe uncertainties to the derived annual emission values.

The emissions of HFCs with short atmospheric lifetimes are systematically underestimated by inverse modeling methods from atmospheric data. As the particle dispersion is modelled backwards in time, some of the HFC can be lost during that transportation time by natural atmospheric degradation. The amount of HFC detected by the measuring station will then be less than the amount emitted at the source. For example, for HFC-152a (comparatively short lifetime, see Table 1) in a 20 day backward simulation this error can lead to a result up to 3.5% below the actual value (Stohl et al., 2009).

In a comparative study of four models based on inversion, Brunner et al. (2017) found that FLEXPART based models yield better results than the NAME-based UKMO system. The different dispersion models, underlying meteorological models and the model setup like the particle release height for the backwards simulation are all factors that could possibly contribute to the performance difference. For the individual country emission estimates the uncertainty was higher for countries further from measurement stations, like Spain and Portugal where uncertainties in the range 42–51% were found, while in the UK this range was 5–22% (Brunner et al., 2017).

The results of the four different models used by Brunner et al. (2017) do not fall within the uncertainty intervals of each other. This mismatch shows that the uncertainty given by the models is smaller than the real uncertainty. Several factors playing a role in creating the differences not covered by the analytical uncertainties were identified. Amongst these

factors are the subsampling of observations used in the model, the treatment of the background and whether the correlation structures of the prior uncertainties are considered, especially the spatial distribution. Likewise, the over- or underestimation of boundary layer heights was mentioned by Stohl et al. (2010) as an example for a systematic error. Recently, a number of atmospheric tracers have been identified, which can be used in the future to assess and improve atmospheric transport models (Simmonds et al., 2020).

Aggregation of emissions to country levels poses a significant error source. Emissions from grid cells containing several countries need to be distributed. This distribution varies in different models and follows either the area fractions covered by the respective countries or the relative share of population within the emission grid cell area (Brunner et al., 2017). However, grid cells oriented at national borders do not necessitate distribution, eliminating this error source. Coarse grids led to total country emission errors of 1–6% (Brunner et al., 2017). The use of a proxy like the night light distribution for population density may contribute to uncertainty added by aggregating or disaggregating emission values, since they might not represent the locations of the emissions. Bottom-up inventories are mostly national reports generated from country level data, which suggests that there could be stronger error correlations within country borders in the prior information (Brunner et al., 2017). However, this approach does not seem to be applied in practice yet.

Typical uncertainty values of inverse modelling emission estimates in top-down studies reviewed here are around 25%–30%, ranging from below 10% up to 90% (considering average, median and range of top-down estimates underlying Fig. 3). The high uncertainty values of above 80% are mostly associated with relatively small emission values.

2.4. Interspecies correlation

Interspecies correlation (ISC) exploits the correlation of the HFC emissions (target compound) with those of a tracer compound. Simmonds et al. (2017) derived US emissions of several HFCs using HFC-125 as a reference and basing their estimate on HFC-125 reported emissions to the UNFCCC. Kim et al. (2010) found significant correlation between HCFC-22 and HFC-143a and derived the emission value of the HCFC-22 tracer by inverse modelling using the FLEXPART transport model. Fang et al. (2012) compared CO and HCFC-22 as tracer molecules and found comparable results in their estimation of Chinese HFC-134a emissions. Halocarbon emissions from the US and Mexico have also been derived from aircraft measurement data by using CO as a tracer molecule (Millet et al., 2009).

3. Bottom-up emission estimation

Bottom-up methods are very heterogeneous, and inventories differ significantly from country to country.

Most bottom-up emission estimates are conducted using the emission-factor approach, as presented in the IPCC guidelines (Ashford et al., 2006). In this approach, emissions are inferred from consumption data and emission factors. This can be performed in an aggregated (IPCC method category Tier 1) or disaggregated (IPCC method category Tier 2) manner, in which general information is considered for more detailed data per sub-application with specific emission factors.

Using a model for refrigeration and air conditioning (RAC) applications, bottom-up estimates of HFC emissions from that sector can be derived. By varying the input factors of the RAC model such as refill, unit lifetime, market penetration (% of cars with AC) and life-cycle emission rates, Say et al. (2016) tried to test the sensitivity of the model and bring the bottom-up estimates in closer agreement with the emissions they derived from atmospheric measurements. Combining a lower refill and market penetration rate can significantly decrease the RAC model emission estimate. This lowered value is, however, still higher than the estimates inferred from atmospheric measurements (Say et al., 2016).

The Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model from the International Institute for Applied Systems Analysis (IIASA) uses emission factors adapted to the country, for example with specific information about maintenance levels or transportation fleet composition (Tohka, 2005). Wherever information is available, emission factors are adjusted for specific HFC emission sources. For each sector, a common GWP is calculated from the shares of the used HFCs and their respective GWPs (CCAC and UNEP, 2016).

The Emissions Database for Global Atmospheric Research (EDGAR) from the European Commission Joint Research Centre (EC-JRC) and the Netherlands Environmental Assessment Agency (PBL) contains bottom-up estimates of HFCs, among other greenhouse gases, on a country level for most countries globally up to 2010 (PBL and EC-JRC, 2017).

In the 2017 UNIDO report on the South African HFC inventory, trade data on HFC import and export was collected from state agencies, official trade statistics and major importers and suppliers and verifications from further stakeholders. This data was used for an bottom-up estimation of South African HFC emissions (UNIDO, 2017). Liu et al. (2019) collected production and consumption data from major room air conditioning manufacturers and air conditioning recycling companies to estimate Chinese annual emissions of R-410a, a blend of HFC-125 (50% w) and HFC-32 (50%w) from 2006 to 2017. Emissions of fluorinated gases including HFCs in Korea were estimated as well based on an industry survey with an IPCCC Tier 1a method, resulting in higher emissions of total fluorinated gases than estimated in the nation inventory report of the Korean government (Jung et al., 2020).

3.1. Error sources and uncertainty of bottom-up estimations

The uncertainty of bottom-up estimates is often relatively high and difficult to quantify due to the many assumptions made. Uncertainty in bottom-up studies is mainly determined by quality and completeness of the information underlying the consumption values: data about import and export and sales of chemicals, or market data about product sales (Ashford et al., 2006). Additional sources of uncertainty lay within the specific emission factors for sub-applications or blends and further assumptions necessary for the calculation. Especially in the disaggregated (Tier 2) form, when emissions are estimated at the sub-application level or even more detailed, the selection of appropriate emission factors is relevant (Ashford et al., 2006). Emission factors have also been identified as a driver for high uncertainty of bottom-up emission estimates for another greenhouse gas, methane, derived with GAINS and EDGAR methodologies (Cheewaphongphan et al., 2019). One of the major obstacles in the collection of data for detailed national inventories (Tier2) are confidentiality issues preventing companies from disclosing unaggregated data (Ashford et al., 2006).

Activity data and leakage rates of cooling equipment contain high uncertainty (CCAC and UNEP, 2016). The uncertainty of emission estimates from different sectors is highest in stationary air conditioning, while commercial refrigeration and mobile air conditioning emission estimates also contain high uncertainty (Purohit and Höglund-Isaksson, 2017).

While emission factors seem to be most important for the accuracy of historical emission estimates, uncertainty in activity data is the largest source of the total uncertainty in emission projections (CCAC and UNEP, 2016). The efficiency of air conditioners, refrigerators and other devices is gradually improving over time and alternatives to HFCs are increasingly used. This might lead to out-of-date assumptions for the bottom-up estimates of emissions from numbers of devices and could be a factor contributing to the overestimation of emissions in bottom-up estimates for some HFCs compared to top-down estimates found in some parts of the world (CCAC and UNEP, 2016).

In the study of R-410 A emissions by Liu et al. (2019), uncertainties for the bottom-up estimations were analyzed by Monte-Carlo simulations. They furthermore showed that almost all emissions occur at the end-of-life stage of room air conditioners. This highlights the great need

for proper disposal and handling of recycling processes. Waste, and especially electronic waste, of which some is containing HFCs (e.g. refrigerators), is often exported (LaDou and Lovegrove, 2008). While the export may be better recorded, it may be that imports into waste processing countries are not accounted for in bottom-up estimates. Many waste streams lead from Annex I to non-Annex I countries (Sthiannopkao and Wong, 2013). Currently, the emissions from that waste stream are hence not included in the inventories used to estimate emissions reported to the UNFCCC, because exports of waste are subtracted from national consumption. Since the last stage of the life cycle of products contributes vastly to HFC emissions (Zhao et al., 2015), this could potentially be a relevant factor. As emissions would be accounted for in countries where only the disposal happens but the benefits of the product were not used, this method of accounting raises fairness issues.

The methods used by each country to derive national inventories of HFCs are different, which complicates a comparison between countries. Sometimes inventories are incomplete, e.g. not considering one or several sectors of HFC use (Climate and Clean Air Coalition CCAC, United Nations Environment Programme UNEP, 2016). This incompleteness and the use of general emission factors, which are not adapted to the specific situation of a country puts doubt on the reliability of national emission estimates, which in turn hinders a meaningful comparison (CCAC and UNEP, 2016). These findings for Latin America and the Caribbean can be generalized to the global scale, as harmonization efforts beyond the IPCC guidelines are scarce. Often, emissions of HFCs are aggregated and given in CO_2-equivalents with the result that information on specific gases and sectoral distribution is lost. UNEP publishes only GWP-weighted data in CO_2eq on their website, which makes a comparison more difficult if the exact GWPs used are not provided. Aggregated CO_2eq emissions contain less information about the real amount of emissions, as GWPs of HFCs vary widely and thus the overall mean GWP depends on the mix of gases emitted. Discrepancies of estimates obtained from different methods are then hard to explain and gaps in reporting hard to identify, which hinders control and enforcement of regulations. However, CO_2eq emissions directly show the actual effect and highlight the overall target of lowering human induced radiative forcing.

Most bottom-up studies reviewed here do not give any estimate of the uncertainty contained in the given values. Uncertainties are neither included in bottom-up data reported to the UNFCCC. Most bottom-up studies are inventories published by governments or other organization organizations, hence they are not peer-reviewed as scientific top-down studies are.

4. Comparison of top-down and bottom-up estimations of HFC emissions

4.1. Aggregate HFC emissions

On a global scale, emission estimates reported to the UNFCCC by Annex I parties were found to make up for little more than half of CO_2-eq emissions derived by atmospheric measurements (Montzka et al., 2015; Montzka et al., 2018; UNFCCC, 2020b). Non-Annex I countries of the UNFCCC are not required to report HFC emissions. These parties are usually Article 5 countries in the context of the Montreal protocol, where they are required to report HFC consumption under the Kigali Amendment from 2020 on (Ozone Secretariat, 2018). Fig. 2 shows top-down global total emissions and bottom-up derived emissions from Annex I countries per gas in 2016. When aggregating emissions of all ten displayed gases (in Gg yr^{-1}), UNFCCC reports from Annex I countries account for 40% of atmosphere-derived global HFC emissions in 2016.

Lunt et al. (2015) studied five different HFCs and found that differences between reported and modelled estimates observed for single compounds from one same country or region cancelled out when considering aggregate HFC emissions. For Annex I countries, emissions derived with their inverse modelling approach matched emissions reported to the UNFCCC by these countries when considering the aggregate CO_2eq HFC emissions. This agreement of emission values suggests that the previously found mismatches in reported and top-down modelled emissions are largely due to non-Annex I country emissions (Lunt et al., 2015).

For total HFC emissions, it is estimated that China contributes only about 35% to emissions of HFCs from the non-Annex I group of countries, indicating that other big non-Annex I emitters exists and share responsibility for the gap (Fang et al., 2016; Montzka et al., 2018).

Emissions of HFCs both reported to the UNFCCC and measured by NOAA are increasing (Montzka et al., 2018). However, the gap of reported emissions and measured emissions is increasing as well. A small difference could be explained by emissions from banks, when they are not accounted for accurately in bottom-up estimations (Montzka et al.,

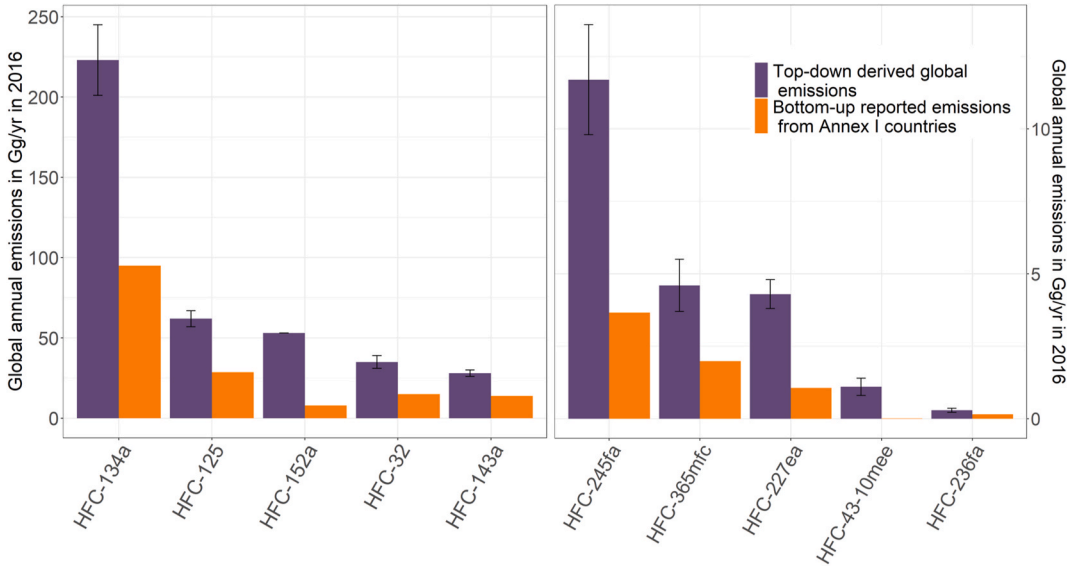


Fig. 2. A comparison of global top-down derived emission estimates (Montzka et al., 2018) and bottom-up estimated emissions reported to the UNFCCC by Annex I countries in Gg yr^{-1} for 2016 per HFC gas (UNFCCC, 2020b). Uncertainty ranges of top-down estimates are 201–245 Gg yr^{-1} for HFC-134a, 57–67 Gg yr^{-1} for HFC-125, 31–39 Gg yr^{-1} for HFC-32, 26–30 Gg yr^{-1} for HFC-143a, 9.8–13.8 Gg yr^{-1} for HFC-245fa, 3.7–5.5 Gg yr^{-1} for HFC-365mfc, 3.7–4.8 Gg yr^{-1} for HFC-227ea, 0.80–1.4 Gg yr^{-1} for HFC-43-10mee and 0.22–0.36 Gg yr^{-1} for HFC-236fa. No uncertainty range was given for HFC-152a.

2015; Velders et al., 2014).

Many studies that conducted atmospheric measurements of HFCs and estimated emissions from a top-down approach using inverse modelling or interspecies correlation compared their findings with bottom-up estimates, either from the EDGAR inventory or the UNFCCC reports of Annex I countries (PBL and EC-JRC, 2017; UNFCCC, 2020a, 2020b). In the following, top-down and bottom-up emission estimates are comparatively discussed per HFC gas. Fig. 3 shows emission estimates of the five most common HFCs for China, Europe, and the USA from top-down and bottom-up studies reviewed here.

4.2. HFC-134a

The emissions of HFC-134a are the fastest growing HFC emissions, reaching 223 (201–245) Gg yr^{-1} in 2016 (Montzka et al., 2018). Top-down studies find both lower and higher emission estimates than bottom-up reports. European average emissions from 2003 to 2014 derived from atmospheric measurements (20.1 ± 6.3 Gg yr^{-1}) are 25% lower than the emissions derived from data reported to the UNFCCC and for more recent years, top-down estimates are even lower (Graziosi et al., 2017; Schoenenberger et al., 2018). This mismatch likely originates from too high emission factors used in the bottom-up calculation. EDGAR shows a yet higher emission estimate (Graziosi et al., 2017). Say et al. (2016) used the NAME model and the inversion technique InTEM from the UK Meterological Office to estimate HFC-134a emissions from the UK from atmospheric measurement data and compared it to UNFCCC reported emissions. They find that reported emissions estimated with the bottom-up RAC model used for UNFCCC reporting are almost twice the magnitude of top-down derived emissions. For Swiss emissions of HFC-134a, too, values where higher in bottom-up reporting than in top-down estimates (Henne et al., 2020).

On the other hand, higher top-down estimates were found for example by Keller et al. (2012) by inverse modelling of HFC-134a emissions than reported from Romania in 2009. Emissions estimated in an inverse modelling study by Fortems-Cheiney et al. (2015) are generally higher than most other top-down estimates, and often also higher than bottom-up estimates. Emissions of HFC-134a from Japan derived by inverse modelling by Fortems-Cheiney et al. (2015) were with 12 Gg yr^{-1} from 2006 to 2008 much higher than reported to the UNFCCC and much higher than other top-down (inversion) estimates would indicate (Li et al., 2011; Lunt et al., 2015; Stohl et al., 2010). Emissions of HFC-134a reported to the UNFCCC by Japan in 2007 were confirmed by these other studies based on inverse modelling. Bottom-up estimates of Japanese emissions from EDGAR, which were used as a priori information, are even higher than the posterior results from Fortems-Cheiney et al. (2015). The authors suggest the choice of the prior data as major influence explaining the different results of the top-down studies. Also for Chinese HFC emissions, estimates by Fortems-Cheiney et al. (2015) for the years before 2010 were higher than other top-down studies (Kim et al., 2010; Li et al., 2011; Stohl et al., 2010; Yokouchi et al., 2006), while their values were closer to bottom-up estimates, except for unrealistically low EDGAR values (Su et al., 2015; UNFCCC, 2020a). A later top-down study by Yao et al. (2019) finds 19 Gg yr^{-1} for 2011, which is closer to the estimate from Fortems-Cheiney et al. (2015) of 20 Gg yr^{-1} for 2010. Likewise, for Europe and the USA, estimations from the inverse modelling study by Fortems-Cheiney et al. (2015) are higher than other top-down estimates (compare Fig. 3).

A top-down study of HFC-134a emissions from the USA based on ISC shows increasing emissions from 2008 to 2012, reaching higher values than reported to the UNFCCC (Simmonds et al., 2015). Hu et al. (2015) used a Bayesian approach to the inversion problem of estimating HFC-134a emissions in the US. Emission estimates of HFC-134a in the US derived from inverse modelling are close to reported emission values, except a decreasing trend from 2010 to 2012 deviating from the reported values which remained steady (Hu et al., 2015, 2017). However, for the most recent years 2013 and 2014 UNFCCC reported emissions are matched.

Comparing atmospheric measurements with UNFCCC reports for HFC-134a on a global scale exposes a big gap. The UNFCCC reported emissions from Annex I countries account for 43% of the emissions derived from measurements globally in 2016 (Fig. 2, see also supplementary data file). This gap in HFC-134a emissions constitutes most of the global gap between reported and measured emissions of all different HFCs together. Due to the probable overestimation of European HFC-134a emissions in the UNFCCC data, the gap is likely even larger.

China is the biggest non-Annex I emitter of HFCs (Montzka et al., 2018). Chinese emissions of HFC-134a in 2016 were estimated to 30 (24–36) Gg yr^{-1} by inverse modelling (Yao et al., 2019). As much as 44% of global HFC-134a emissions are thus left to stem from the group of non-reporting (non-Annex I) countries other than China.

While previous bottom-up estimates for HFC-134a from India were 1.1 Gg yr^{-1} in 2005 (Garg et al., 2006) and 1.67 Gg yr^{-1} in 2007 (Sharma et al., 2011), the first top-down estimation published by Say et al. (2019) found significant emissions of HFCs from India in 2016 (8.2 (6.1–10.7) Gg yr^{-1} for HFC-134a). This large mismatch indicates a very rapid growth in HFC emissions since 2005/2007, which was projected previously (Chaturvedi et al., 2015), and (or) a significant difference between top-down and bottom-up estimations (Say et al., 2019). However, this measured value of HFC emissions from India can only explain a small fraction of the 44% (about 98 Gg yr^{-1}) gap in global emissions in 2016, which is attributed to all non-Annex I countries, excluding China.

Consumption data can give some indication about possible emissions and are especially interesting from non-Annex I countries, which are not reporting emissions to the UNFCCC. HFC-134a consumption estimations from non-Annex-I countries Chile (Ministry of Environment Chile, 2014), Colombia (UTO and UNDP, 2014), Ghana (Ashford, 2016b), South Africa (UNIDO, 2017), Liberia (EPA Liberia, 2017), Bangladesh (Reazuddin, 2014), Vietnam (VNEEC, 2017), Indonesia (Pasek, 2014), Moldova (Ashford, 2016c) and Jordan (UNIDO, 2018) are available. Each report mentions consumptions below 5% of the Chinese consumption for 2012 or 2013. These countries are thus likely not significant in explaining the gap (Figure S1).

4.3. HFC-32

UNFCCC reported emissions of HFC-32 from Annex I countries were 43% of global emissions derived from atmospheric measurements in 2016 (Fig. 2). According to top-down estimates for 2016, emissions from China can explain 50% of this emission gap (Fang et al., 2016; Lunt et al., 2015; Yao et al., 2019), while for earlier years it has been described that Chinese emissions could explain most of the gap (Fang et al., 2016; Lunt et al., 2015; Montzka et al., 2018). However, UNFCCC reported emissions from Europe could be higher than actual emissions, widening the gap again. HFC-32 emissions of 2.3 ± 0.8 Gg yr^{-1} in Europe were estimated by inverse modelling for the years 2003–2014 on average (Graziosi et al., 2017). While this emission value agrees with UNFCCC data, the annual growth rates are substantially higher in UNFCCC data (2.6% growth rate from inversion model, 13.3% in UNFCCC data). For 2013, 2014, the most recent years considered in the top-down study, UNFCCC reported emissions are about 50% higher than the top-down estimates. EDGAR estimates were much lower, probably due to incomplete inventory data (Graziosi et al., 2017). The emissions of HFC-32 from the USA were lower in the UNFCCC reported estimates than in estimates derived from atmospheric measurements for earlier years, but generally matched quite well (Hu et al., 2017; Simmonds et al., 2015; UNFCCC, 2020b).

Indian national emissions could not be inferred from atmospheric measurement campaigns, because measured HFC-32 concentrations were expected to originate from production sites rather than from products and therefore not considered to scale with population density (Say et al., 2019).

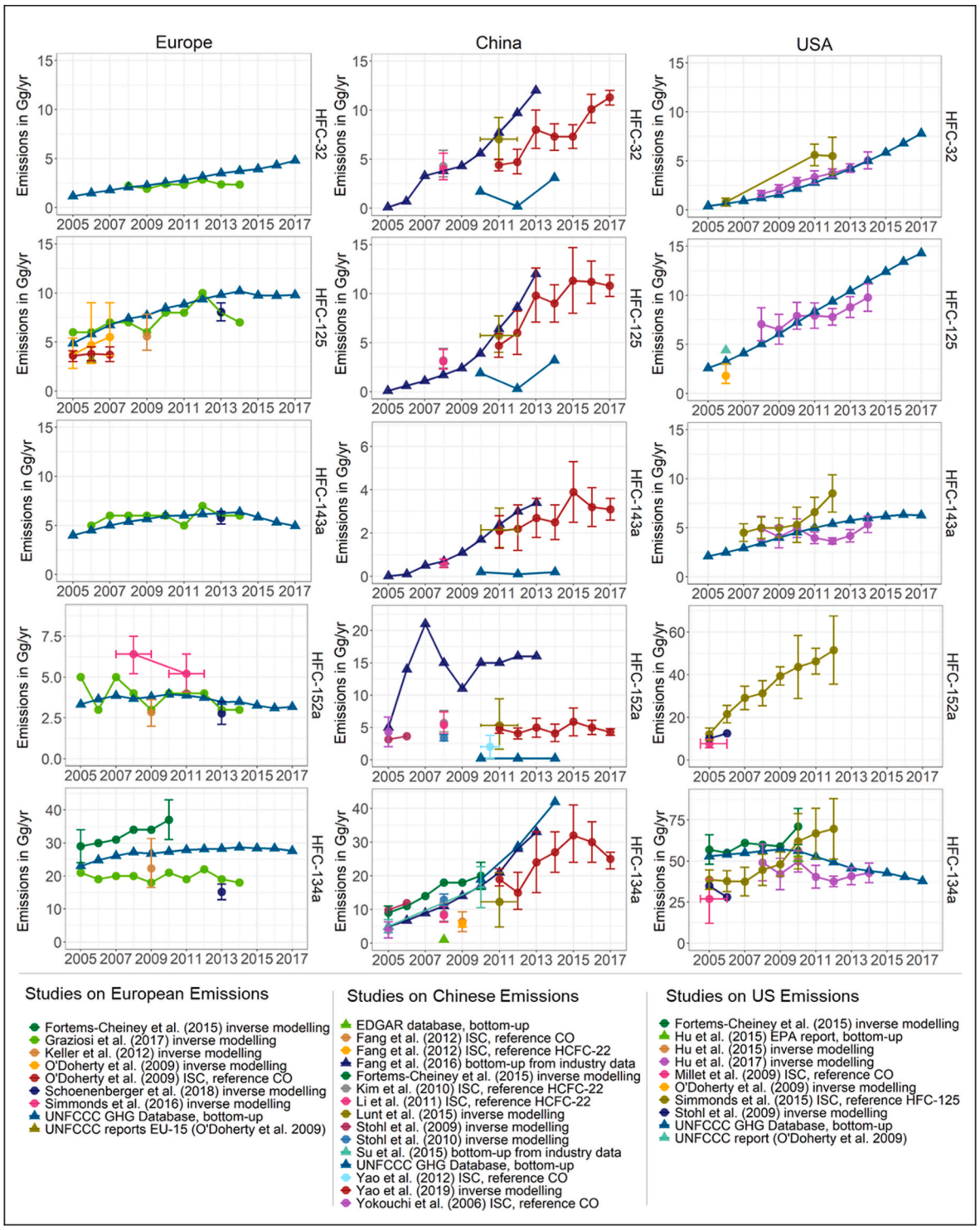


Fig. 3. Emissions of HFC-32, HFC-125, HFC-143a, HFC-152a and HFC-134a from China, the USA and Europe between 2005 and 2017 according to different studies. Bottom-up estimates are marked with triangles and top-down results with dots. X-error bars indicate time ranges of more than one year, for which average values were given in the respective study. In the ISC study by Simmonds et al. (2015) no values with $R^2 > 0.5$ were obtained for HFC-32 for 2007–2010, thus they are not included here. The studies on European emissions used different geographical boundaries. Graziosi et al. (2017) did not give uncertainty intervals for their emissions estimates for the total European Geographical Domain in the supplementary data tables, therefore no error bars are displayed. Uncertainty for regional estimates varied between 15% and 83% for HFC-32, 16% and 80% for HFC-125, 20% and 82% for HFC-143a, 19% and 80% for HFC-152a and 12% and 80% for HFC-134a. Values from Schoenenberger et al. (2018) for Europe were obtained by subtracting emissions estimated for subregions Maghreb, Turkey, Egypt and Middle East from the emission estimate for the total domain of the study. (Fang et al., 2012; Fang et al., 2016; Fortems-Cheiney et al., 2015; Graziosi et al., 2017; Hu et al., 2015; Hu et al., 2017; Keller et al., 2012; Kim et al., 2010; Li et al., 2011; Lunt et al., 2015; Millet et al., 2009; O'Doherty et al., 2009; Schoenenberger et al., 2018; Simmonds et al., 2015; Simmonds et al., 2016; Stohl et al., 2009; Stohl et al., 2010; Su et al., 2015; UNFCCC, 2020a, 2020b; Yao et al., 2012; Yao et al., 2019; Yokouchi et al., 2006).

4.4. HFC-125

46% of global atmospheric measurement-derived emissions of HFC-125 in 2016 were matched by UNFCCC reported emissions from Annex I countries (Fig. 2). Emission estimates from China can explain one third of this emission gap for HFC-125 in 2016 (Yao et al., 2019), leaving about 22 Gg yr^{-1} to stem from other non-Annex I countries or method discrepancies.

US emissions of HFC-125 reported to the UNFCCC are slightly higher than estimates derived from atmospheric measurements by inverse modelling, which show emissions of 9.77 (8.37–11.17) Gg yr^{-1} in 2014 (Hu et al., 2017). For HFC-125 from Europe, yearly average emission of 6.9 ± 2.4 Gg yr^{-1} were found for 2003–2014, where EDGAR and the UNFCCC estimates lay within the uncertainty interval, but are also on the higher end (Graziosi et al., 2017). Keller et al. (2012) found much higher emissions of HFC-125 in 2009 than reported from Poland. Australian HFC-125 emissions reported to the UNFCCC were up to one third higher than top-down estimates from ISC and inverse modelling, which estimated 0.60 (0.40–0.79) Gg yr^{-1} and 0.74 (0.49–0.99) Gg yr^{-1} for 2016, respectively (Dunse et al., 2018).

Emissions from India in 2016 were 6.4 (5.2–7.8) Gg yr^{-1} according to inverse modelling results, explaining about 30% of emissions from non-Annex I countries other than China (Say et al., 2019). The authors ascribe difficulties of the inversion model to capture HFC-125 emission elevations over the baseline to point sources. HFC-32 and HFC-125 emissions often correlate, indicating that emissions resulted from the use of the refrigerant blend R-410 A (50 wt% HFC-125, 50 wt% HFC-32) (Say et al., 2019).

4.5. HFC-143a

For HFC-143a, 50% of global emissions derived from atmospheric measurements were matched by UNFCCC reported emissions from Annex I countries in 2016 (Fig. 2). Emission estimates from China account for 11% of total global emissions of HFC-143a (Yao et al., 2019), resulting in about 11 Gg yr^{-1} emissions to stem from other countries.

Emissions of HFC-143a from the EU and USA reported to the UNFCCC are matched quite well by top-down estimates (Graziosi et al., 2017; Hu et al., 2017; Montzka et al., 2018; Schoenenberger et al., 2018; UNFCCC, 2020b). HFC-143a emissions contributed the most to European CO_2eq emissions and per capita emissions in Europe are more than four times larger than average global per capita emissions (Graziosi et al., 2017). Australian UNFCCC reported emissions reached more than double the values derived by top-down methods, which estimated HFC-143a emissions below 0.5 Gg yr^{-1} (Dunse et al., 2018). Inverse modelling found HFC-143a emissions below 1 Gg yr^{-1} from India in 2016 (Say et al., 2019), which are less significant compared to the unexplained remainder of the gap.

4.6. HFC-152a

Reported emissions from Annex I countries to the UNFCCC account for only 15% of global emissions derived from atmospheric measurements in 2016 (Fig. 2). The emissions of HFC-152a from the USA are reported only aggregated with other F-gases to the UNFCCC, because of confidentiality issues (Montzka et al., 2018). The majority of the gap can be explained by US emissions, which is supported by atmospheric measurements and subsequent inversion to derive emissions of HFC-152a from the USA (Simmonds et al., 2015, 2016). Chinese HFC-152a emissions were estimated to 5 (3.9–6.1) Gg yr^{-1} (Yao et al., 2019), which represents 9.4% of global emissions.

Emissions of HFC-152a in Europe derived by inverse modelling were 4.1 ± 1.8 Gg yr^{-1} on average during the period 2003–2014 and largely agree with UNFCCC reported emission estimates (Graziosi et al., 2017). Another inverse modelling study found much higher emission estimates (Simmonds et al., 2016). Both inversion studies show a decreasing trend. Emissions of HFC-152a from southeast and central south Europe were reported to decrease significantly, but the top-down estimate performed by Keller et al. (2012) could not confirm this trend. Their emission results are much higher than reported values.

Difficulties were encountered in inverse modelling of HFC-152a emissions from Turkey due to temporal variabilities (Schoenenberger et al., 2018). The authors suspect a point source of emissions such as a factory to be the cause.

Bottom-up reported estimates of HFC-152a for India are much smaller than estimates derived from atmospheric measurements (Say et al., 2019). However, Indian emissions only contribute 2–3% to global HFC-152a emissions.

4.7. HFC-227ea

UNFCCC reported emissions of HFC-227ea from Annex I countries make up 25% of global top-down derived emissions (Fig. 2). Chinese emissions can account for 26% of global HFC-227ea emissions in 2016, explaining a third of the gap (Yao et al., 2019). With European emissions of 0.41 ± 0.22 Gg yr^{-1} for HFC-227a, the inversion model result shows high similarity to the UNFCCC estimates (Graziosi et al., 2017). The authors found that EDGAR estimates for HFC-227ea are much higher and show an increasing trend of emissions, which was not found in the inversion model results. Graziosi et al. (2017) state that the uncertainty of HFC-227a measurements in the atmosphere is high due to its lower concentration compared to other F-gases, whereas Brunner et al. (2017) found measurement errors to play a minor role in uncertainties of inversion estimates in their analysis.

4.8. HFC-236fa

For HFC-236fa, 52% of global emissions derived from atmospheric measurements are accounted for in UNFCCC reports from Annex I countries (Fig. 2). Inverse modelling shows emissions of HFC-236fa from China made up 35% of global emissions in 2016 (Yao et al., 2019), leaving a remaining gap of 13% or 0.04 Gg yr^{-1}. For Europe, HFC-236fa emissions of 0.02 ± 0.01 Gg yr^{-1} with a positive trend of 3.6% annually were estimated by inversion (Graziosi et al., 2017). As for HFC-227a, high measurement uncertainty is contained in this result. For HFC-236fa, the inversion shows emissions larger by 50% than the estimates of UNFCCC and EDGAR, indicating a gap in reporting (Graziosi et al., 2017). Vollmer et al. (2011) also find a significant underestimation of reported data when comparing the values to their results derived from a global inversion model.

4.9. HFC-365mfc

UNFCCC reported emissions of HFC-365mfc from Annex I countries account for 43% of top-down estimated global emissions (Fig. 2). Chinese emissions of HFC-365mfc were estimated by inverse modelling to 0.4 (0.3–0.6) Gg yr^{-1} in 2016 (Yao et al., 2019), leaving a remaining gap of 48% of global emission. Industrial areas like the Po Valley in Italy were identified as major sources of emission of HFC-365mfc and Europe was the dominant source of HFC-365mfc emissions in the first decade of the century (Stemmler et al., 2007; Vollmer et al., 2011). Bottom-up estimates for Europe showed considerable underestimation, which could be responsible for part of the gap. Inverse modelling estimated average European emissions of 1.2 ± 0.6 Gg yr^{-1} from 2005 to 2014, which is three times as much as emissions reported to the UNFCCC (Graziosi et al., 2017). HFC-365mfc was the only HFC studied by Graziosi et al. (2017) that was significantly underestimated by bottom-up methods (UNFCCC, EDGAR) compared to their inversion results. As HFC-227a and HFC-236fa, also HFC-365mfc is present in the atmosphere in very low concentrations, leading to higher measurement uncertainties (Graziosi et al., 2017).

4.10. HFC-245fa

31% of global top-down derived emissions of HFC-245fa are met by UNFCCC reports from Annex I countries for 2016 (Fig. 2). Chinese emissions in 2016 made up 10% of total global emissions (Yao et al., 2019). The remaining gap of HFC-245fa emissions is 6.84 Gg yr^{-1}. HFC-245fa is predominantly consumed in North America, where it is used as a foam blowing agent for polyurethanes (Graziosi et al., 2017). HFC-245fa emissions from Europe over the period 2008–2014 were estimated at 0.74 ± 0.33 Gg yr^{-1} with a decreasing trend. UNFCCC data is in agreement, while EDGAR estimates a strong positive trend (Graziosi et al., 2017).

4.11. HFC-43-10mee

Less than 1% of global emissions of HFC-43-10mee derived from atmospheric measurements are matched by bottom-up reports to the UNFCCC from Annex I countries (Fig. 2). HFC-43-10mee is mainly used as a cleaning agent in the electronics industry (Arnold et al., 2014). Emissions have been stagnating since 2007 (Arnold et al., 2014; Montzka et al., 2018). Confidentiality issues could be a reason for low reporting.

5. Measurement stations coverage and top-down studies distribution

Stohl et al. (2009) note that the northern hemisphere is covered much better by networks of atmospheric measurement sites than the southern hemisphere, resulting in difficulties to quantify emissions especially from South America and Africa as well as India, Indonesia and northern Australia due to low sensitivities (compare Fig. 1). Indonesia, Brazil, Iran and Saudi Arabia are among the highest overall greenhouse gas emitters (Crippa et al., 2019). Therefore, top-down estimations from these countries would be very interesting. Further top-down estimations would also help to quantify HFC emissions from India and Mexico, which both have high population numbers and greenhouse gas emissions. Bottom-up estimations for the mentioned countries are scarce as well, but the assessment of HFC emissions from these Article 5 states would help to explain the gap in emissions between reported values from Annex I countries to the UNFCCC and global emissions derived from atmospheric measurements.

Although there are several permanent stations on the comparatively small continent (see Fig. 1B), Brunner et al. (2017) found that more atmospheric measurement stations in Europe are necessary to geographically restrain the emissions obtained from inverse modelling to the country level. National estimates in Europe pose a challenge, as countries are small. Especially in eastern Europe the sensitivity of the current measurement stations is not high enough for a reliable country-level resolution of top-down estimates (Brunner et al., 2017). Emission estimates for Spain and Portugal depended strongly on the prior information given to the model, demonstrating that the network of the three stations Monte Simone, Jungfraujoch and Mace Head fails to provide a sufficient sensitivity for robust emission estimates from the Iberian Peninsula (Brunner et al., 2017).

Much higher sensitivities for eastern European HFC emissions were achieved when including a campaign measurement site in K-Puszta, Hungary next to the stations in Italy, Switzerland, and Ireland (Brunner et al., 2017). The strategically selected measuring location in Hungary not only made top-down emission estimates for eastern Europe possible but also helped to better constrain central European emission estimates (Keller et al., 2012).

The addition of data from the Finokalia measurement station in Greece reduced uncertainty in inverse modelling emission estimations by 40–80% for the Eastern Mediterranean region and by 400% for the national emission estimate for Greece (Schoenenberger et al., 2018). A permanent measuring station at this site would also allow to provide emission estimates for Northern Africa and the Middle East using transport events from that region.

Mt. Mugogo in Rwanda is a relatively new station within the AGAGE network which is equipped with Medusa MS system (MIT and NASA). So far, only data on carbon dioxide and methane concentrations were analyzed, but no analysis of HFC measurements was published from this site. Measurement data of HFCs from this site would be interesting for estimations of emissions from African countries like Nigeria, Kenya and Tanzania, for example. Possibly, emissions from Saudi Arabia and other countries on the Arabian Peninsula could be estimated with data from Mt. Mugogo as well, which would be of high interest because to date very limited information is available for this region. Furthermore, GDP and overall greenhouse gas emissions of those countries are quite high, indicating a potential for high HFC emissions (World Bank, 2019). The feasibility of emission estimates based on data from very distant measuring sites was demonstrated previously by estimating US emission with measurement data from Mace Head, Ireland (Simmonds et al., 2015). However, the occurrence of suitable atmospheric transport events must be evaluated.

The ENEA Station for Climate Observations on the island of Lampedusa, Italy, will also start including HFCs into their measurements (ENEA, 2010–2020). These measurement data points could be valuable as well to better quantify emissions in the Mediterranean and possibly the Middle East.

The global distribution of top-down studies resembles the distribution of measurement stations. For Latin America and the Caribbean, only one top-down emission estimation (HFC-134a, from Mexico) was found. Also, for Africa, West Asia and India very few top-down studies were conducted (Fig. 4).

6. Situation in different parts of the world

The global distribution of HFC emissions is quite inhomogeneous. Canada, Japan, Australia, and Russia account for about 20% of HFC emissions reported from Annex I countries, while the majority (about 80%) stem from the US and the EU (Montzka et al., 2018). The situation in different parts of the world is very different also regarding the availability of information on HFC emissions. In the following, data availability and emissions of the five most important HFCs are discussed per world region.

6.1. Africa

The measurement station at Cape Point was used to quantify 2016 emissions of HFC-125 (0.8 (0.5–1.2) Gg yr^{-1}) and HFC-152a (1.1 (0.6–1.6) Gg yr^{-1}) from South Africa, the biggest overall greenhouse gas emitter in Africa, which is 1% and 2% of total global emissions, respectively (Crippa et al., 2019; Kuyper et al., 2019). HFC emission estimates for the Maghreb region and Egypt were included in a top-down study on the Mediterranean (Schoenenberger et al., 2018). For several African countries, such as Ghana and Liberia bottom-up estimations of HFC consumption or emissions exists, prepared by NGOs or government agencies (Ashford, 2016b; EPA Liberia, 2017; Ashford, 2016d; Omotosho, 2015).

6.2. Asia

Many studies investigate HFC emissions from China, employing both top-down and bottom-up methods (see Fig. 3).

HFC-32 and HFC-125 emissions correlated, which reflects the employment of R-410 A as refrigerant in new room air conditioners since approximately 2005 (Yao et al., 2019). China's total HFC production in 2009 was 159 Gg yr^{-1} and aggregate consumption 67.3 Gg yr^{-1} in the same year (Zhang and Wang, 2014). In 2013, production of the five most common HFCs totaled 315 Gg yr^{-1} and their consumption was estimated to 145.4 Gg yr^{-1}, illustrating rapid growth (Fang et al., 2016). Emissions

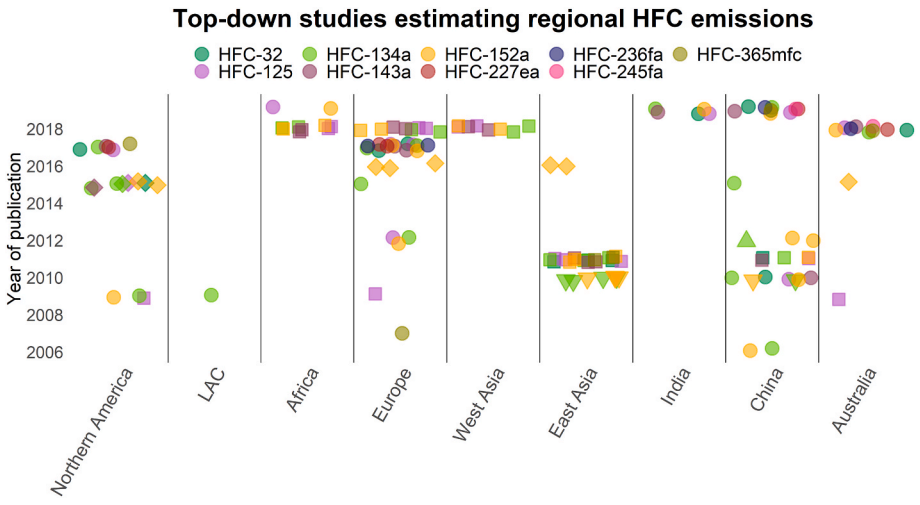


Fig. 4. Overview of gases for which top-down emission estimates were made per world region of measurement and year of publication. Countries and regions for which measurement-based emission estimations were made are divided in the following world regions: Northern America (USA, North America Annex I), Latin America and the Caribbean (LAC) (Mexico), Africa (South Africa, Maghreb, Egypt), Europe (European Geographical Domain, Europe, EU, Balkans), West Asia (Turkey, Middle East), East Asia (Japan, Taiwan, South Korea, North Korea, East Asia), China, India and Australia. Every point represents a country or regional emission estimation of one gas, which is indicated by color. In areas of high overlap different shapes show estimates from different publications. For Northern America and Europe, predominantly recent publications were selected even though more publications from earlier years exist. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

of HFCs are not distributed equally in China. Per capita emissions in Beijing are higher than in mostly rural areas of China (Stohl et al., 2010). Recently, more regional top-down studies of emissions were published, for example form the Yangtze River Delta and the Greater Pearl River Delta region (Pu et al., 2020; Zeng et al., 2020).

Say et al. (2019) derived the first top-down emission estimates of HFCs of India using measurement data from air samples collected by aircraft in June and July 2016. Emissions were obtained by inverse modeling. Accordingly, HFC-134a and HFC-125 constitute the major part of Indian HFC emissions, representing about 4% and 10% of global emissions of these HFCs, respectively (Say et al., 2019).

Aggregated HFC emissions derived by Say et al. (2019) with atmospheric measurements in India for 2016 were an order of magnitude higher than projected by Velders et al. (2015) for that year. As some common refrigerant blends based on HFCs which are widely used as substitutes for CFCs and HCFCs in other parts of the world are apparently not widespread in India according to 2016 measurements, there is a big chance for GHG emission mitigation by directly switching to low-GWP alternatives instead of transitioning through the intermediate use of HFCs (Say et al., 2019).

Top-down studies quantified emissions from other East Asian countries, such as South Korea and Japan (Fortems-Cheiney et al., 2015; Li et al., 2011; Lunt et al., 2015; Stohl et al., 2009, 2010). Japanese reported emissions in 2008 were lower than results of different atmospheric modelling studies indicated. Top-down estimates of HFC-134a ranged from 3.1 (2.9–3.3) Gg yr^{-1} (Stohl et al., 2010) and 4.7 (4.5–5) Gg yr^{-1} (Li et al., 2011) to 12 (10–14) Gg yr^{-1} (Fortems-Cheiney et al., 2015).

Russia's bottom-up estimated emissions reported to the UNFCCC for 2017 are 0.69 Gg yr^{-1} of HFC-32, 1.58 Gg yr^{-1} of HFC-125, 3.91 Gg yr^{-1} of HFC-134a, 0.86 Gg yr^{-1} of HFC-143a, and 0.38 Gg yr^{-1} of HFC-152a. Top-down estimates of HFC-134a and HFC-152a emissions from Russia for the year 2005 and 2006 were three to eight times higher than emissions reported to the UNFCCC for those years (Stohl et al., 2009). Reported emissions of HFC-134a from Turkey were two times higher than emissions estimated by inverse modelling for Turkey and Cyprus in 2013 to 1.42 (0.86–1.97) Gg yr^{-1}, while HFC-125, HFC-143a HFC-152a reported emissions could be confirmed (Schoenenberger et al., 2018).

Further possibly relevant countries on the Asian continent in terms of HFC-emissions considering their population and/or total greenhouse gas emissions, could be the Philippines, Thailand, Indonesia, Vietnam, Bangladesh, Pakistan, Iran and Saudi Arabia (Crippa et al., 2019). For Vietnam, Indonesia and Bangladesh, HFC inventories were prepared as part of a program of the Climate and Clean Air Coalition (Ashford, 2016a; Pasek, 2014; Reazuddin, 2014; VNEEC and CCAC, 2017).

6.3. Australia

In 2016, Australian HFC-134a emissions derived by top-down methods were 1.66 ± 0.55 Gg yr^{-1} (ISC, CO reference gas) and 1.85 ± 0.60 Gg yr^{-1} (inverse modelling) in 2016 (Dunse et al., 2018). UNFCCC reported emissions of HFC-134a in 2016 were 2.9 Gg yr^{-1} (UNFCCC, 2020b). Overall HFC emissions were 50% lower in top-down than in bottom-up methods (Dunse et al., 2018).

6.4. Europe

Top-down and bottom-up estimates of European emissions of the five most common HFCs are shown in Fig. 3. Top-down emission estimates are produced for geographic domains wherein countries are aggregated. Especially in eastern and central Europe, geographic domains for the emission estimation are chosen very inconsistently, which makes it hard to compare results from different studies, as shown in Fig. 5.

For the continent, emission estimates by inverse modelling from Graziosi et al. (2017) and Schoenenberger et al. (2018) are very close to each other, when subtracting emissions estimated for sub-regions Maghreb (Morocco, Algeria, Tunesia, Libya and Mauritania), Turkey, Egypt and Middle East from the emission estimate for the total region considered by Schoenenberger et al. (2018) (see Fig. 3). The region considered by Graziosi et al. (2017) is then, however, larger by the Baltic and Scandinavian states which were not included in the other study.

European emission estimates of HFC-125, HFC-134a and HFC-152a derived from atmospheric measurements using inversion by Keller et al. (2012) are similar to UNFCCC reported values. However, the spatial distribution of the emissions calculated proved to be different from nationally reported emissions. The median of emissions estimated for European countries by all four models compared by Brunner et al. (2017) is 24% higher than the total HFC emissions reported to UNFCCC. Graziosi et al. (2017) found a difference of only 13% between their inversion-based results and reported emissions to the UNFCCC when looking at total HFC emissions in CO_2eq. from Europe. For individual compounds, the different emission estimation methods resulted in larger discrepancies.

6.5. Northern America

Emission estimates of the five most common HFCs from the USA, according to top-down studies and bottom-up reports, are depicted in Fig. 3. US top-down estimates largely confirmed bottom-up results reported to the UNFCCC by the US EPA (Hu et al., 2015, 2017). For HFC-134a and HFC-143a, top-down results from Simmonds et al. (2015)

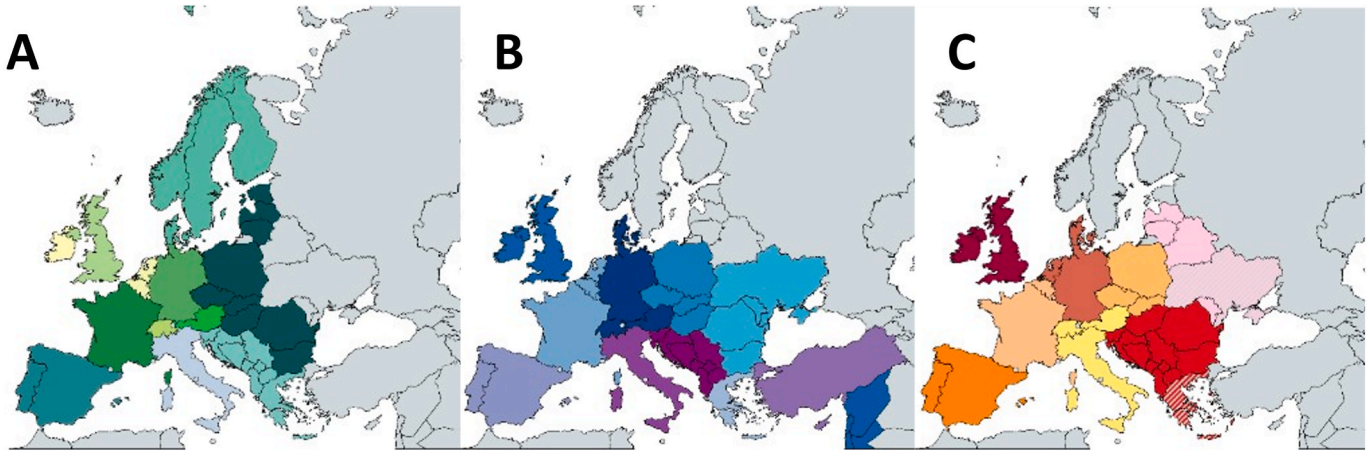


Fig. 5. Different top-down studies on HFC emissions in Europe estimated emissions for different geographical domains. A. Graziosi et al. (2017) (green) grouped emissions in the regions FR (France); UK (United Kingdom); ES-PT (Spain, Portugal); IT (Italy); DE (Germany); NEE (Poland, Czech Republic, Slovakia, Lithuania, Latvia, Estonia, Hungary, Romania, Bulgaria); SCA (Norway, Sweden, Finland, Denmark); SEE (Slovenia, Croatia, Serbia, Bosnia-Herzegovina, Montenegro, Albania, Greece); BE-NE-LU (Belgium, The Netherlands, Luxembourg), IE (Ireland); AT (Austria); CH (Switzerland). B. Schoenenberger et al. (2018) (blue) separated Europe in the regions Turkey (Turkey, Cyprus), Balkans (Serbia, Montenegro, Kosovo, Albania, Bosnia and Herzegovina, Croatia, Slovenia, FYROM), Eastern (Ukraine, Romania, Moldova, Bulgaria), Middle East (Jordan, Lebanon, Syria, Palestine, Israel), Maghreb (Morocco, Algeria, Tunisia, Libya, not colored), Central E (Poland, Slovakia, Czech-Republic, Hungary), Central W (Switzerland, Liechtenstein, Germany, Austria, Denmark), Western (France, Luxembourg, Netherlands, Belgium), Iberian Peninsula (Spain, Portugal) and British Isles (Ireland, UK), Turkey (Turkey, Cyprus) and Greece, Egypt and Italy as national states.C. Keller et al. (2012) (red) considered the following groups: central west (Belgium, France, and Luxembourg), central north (Denmark, Germany, and The Netherlands), northwest (Ireland and the United Kingdom), central south (Austria, Italy, and Switzerland), southeast (Albania, Bulgaria, parts of Greece, Hungary, Romania, and former Yugoslavia), northeast (Czech Republic, Poland, and Slovakia), east (Belarus, Latvia, Lithuania, Moldova, and the western part of the Ukraine), and southwest (Portugal and Spain). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

and Hu et al. (2017) differ in both magnitude and trend.

US HFC-152a emissions are not reported to the UNFCCC, due to confidentiality issues. They were quantified by top-down studies to 7.6 (5.7–9.7) Gg yr^{-1} from 2004 to 2006 by ISC using CO as a reference (Millet et al., 2009) and to 12.1 (9.2–15) Gg yr^{-1} in 2005 by ISC using HFC-125 as a reference (Simmonds et al., 2015). An inverse modelling study finds a value in between, estimating 10.1 Gg yr^{-1} for 2005 (Stohl et al., 2009). Estimates by Simmonds et al. (2015) show a strong increase in HFC-152a emissions, reaching 51.5 ± 16.0 Gg yr^{-1} in 2012, which is vastly more than the highest estimates of HFC-152a emissions from China or Europe (compare Fig. 3).

Canadian bottom-up emission estimates reported to the UNFCCC show an increasing trend for HFC-32, HFC-125 and HFC-143a, while HFC-152a and HFC-134a emissions decrease slightly from 2016 to 2017 (UNFCCC, 2020b). For 2006, Canadian emissions of HFC-134a were estimated by inverse modelling to 3.50 Gg yr^{-1} and emissions of HFC-152a to 1.59 Gg yr^{-1}, which is almost double and more than four-fold the reported values, respectively (Stohl et al., 2009).

6.6. Latin America and the Caribbean

In Latin America and the Caribbean, 80% of HFC emissions are emitted by Argentina, Brazil and Mexico (CCAP and UNEP, 2016). In Brazil, no HFCs are produced (Azevedo et al., 2018). Emissions of HFC-134a according to government reports to the UNFCCC reached 3.9 Gg yr^{-1} in 2015, while HFC-152a were reported to be 0 Gg yr^{-1} in 2007–2015 (Ministério Da Ciência, Tecnologia E Inovação, 2017).

HFC-134a emissions from Mexico in 2006 were estimated to 2.9 (1.7–4.3) Gg yr^{-1} by ISC using CO as tracer molecule (Millet et al., 2009). According to a bottom-up estimation from the Mexican government, 2017 emissions were 0.01 Gg yr^{-1} for HFC-125, 5.15 Gg yr^{-1} for HFC-134a and 3.38 Gg yr^{-1} for HFC-152a, when recalculating CO_2-equivalents to Gg with GWPs shown in Table 1 (Gobierno de México, 2018).

For bottom-up studies, country specific emission factors are often lacking in Latin America and the Caribbean (CCAC and UNEP, 2016). Governmental agencies of some South American countries like Colombia and Chile prepared voluntary reports with bottom-up estimations of HFC consumption (Ministry of Environment Chile, 2014; UTO and UNDP, 2014).

7. Hydrofluoroolefines (HFOs)

Hydrofluoroolefines (HFOs) are one of the replacement options for HFCs. The double bonds in these compounds lead to higher reactivity in the atmosphere (Tovar et al., 2014). The atmospheric lifetimes of HFOs are with 5–22 days much lower than those of HFCs, resulting in lower GWPs of <1 to 6 (Montzka et al., 2015). HFO-1234yf and HFO-1234ze (E) (sometimes referred to as HFC-1234yf and HFC-1234ze(E), both GWP <1 RTOC, 2019) are two prominent examples of HFOs which are already used as substitutes for HFC-134a in different applications (Henne et al., 2012; Montzka et al., 2018; Papasavva et al., 2009). Atmospheric concentrations of HFO-1234yf and HFO-1234ze(E) reached measurable levels in the atmosphere and were detected in 2014 at the remote Jungfraujoch measurement station and an urban site in Switzerland (Vollmer et al., 2015). Consumption of HFC-1234yf in South Africa jumped up from 1 t in previous years to 80 t in 2016, demonstrating that the HFC-alternative is finding its way into the market (UNIDO, 2017).

On one hand, the higher reactivity of HFOs leads to a shorter lifetime and lower GWP of HFOs, which is beneficial for the use as HFC replacements in order to reduce radiative forcing. On the other hand, the atmospheric degradation of HFOs can result in the formation of ozone, which contributes to ground-level pollution, especially in urban areas (Luecken et al., 2010). However, as found for the USA, HFO-1234yf contribution to ozone formation is expected to be low compared to other volatile organic compounds (Luecken et al., 2010; Papasavva et al., 2009).

As for HCFCs and HFCs, one possible degradation product of HFOs is trifluoroacetic acid (TFA) (Solomon et al., 2016; Wang et al., 2018). For HFO-1234fy and HFO-1234ze the molar yield under typical conditions are 100% and 10%, respectively (Solomon et al., 2016). TFA

accumulates in the environment, is toxic to many organisms and can in principle lead to acidification of water bodies (Berends et al., 1999; Lindley et al., 2019). Environmental concentrations of TFA from HFCs and their replacements in the future have been projected to not pose serious threats to environmental or human health (Luecken et al., 2010; Solomon et al., 2016). However, a recent analysis of Arctic ice cores shows that short-chained perfluoroalkylcarboxilic acids, such as TFA, resulting from the degradation of HFCs and other CFC replacements persist in remote areas (Pickard et al., 2020). These persistent and mobile compounds have been identified as reason for concern, as they lead to irreversible contamination (Cousins et al., 2019).

Up to 0.4 °C of global warming by the end of the century can be avoided by the phase-down of HFCs. The same amount of warming could additionally be avoided by efficiency gains in the cooling and air conditioning sectors (TEAP, 2019; Velders et al., 2015). A bottom-up study shows, that the total global warming footprint of the air conditioning and refrigeration sector in India could be reduced by 37% through energy efficiency gains and the use of low-GWP alternatives to HFC refrigerants (Chaturvedi and Sharma, 2015). Another study, investigating the impacts of the Kigali Amendment in Asia, finds that about 10% of the total greenhouse gas emission savings from an HFC phase-down could be attributed to energy savings from synergy effects when installing new equipment (Purohit et al., 2018). On a global scale, the technical potential for co-benefit efficiency improvements in cooling of a full implementation of the Kigali Amendment was estimated to a 20% reduction of global electricity consumption (Purohit et al., 2020). Upon substitution of HFCs with lower GWP alternatives, the overall life-cycle energy efficiency of the refrigerant is thus important to consider (Velders et al., 2012, 2015). Additional safety and environmental concerns beyond global warming warrant a thorough analysis of HFC alternatives, which will be phased-in in the coming years.

8. Conclusions

8.1. Comparability of data

In general, obtaining a complete global overview of all HFC emission data is quite difficult due to inconsistent reporting and the one-off nature of atmospheric studies, providing estimates only for a certain gas in a certain area for a few years. Bottom-up data is often more continuous but more outdated than top-down studies and rarely quantifies uncertainties. The spatial resolution of emission estimates is relevant to be able to compare top-down emission estimates with bottom-up reports at a national scale. A country-level resolution of emission data furthermore enables the control of emission legislation and the evaluation of policy tools. The provision of unaggregated data of individual gases reported in mass and presented in reusable formats could increase transparency and ease comparison.

8.2. Gap between bottom-up and top-down derived emission estimates

Top-down studies on Annex I country HFC emissions, mainly for the EU, USA and Australia, confirmed the general results of bottom-up estimates reported to the UNFCCC to the extent, that the difference to atmospherically derived global emissions could be ascribed mainly to non-Annex I countries and not to large scale underestimation from bottom-up methods.

Numerous studies on Chinese HFC emissions share the conclusion that China is not the only big non-Annex I emitter causing the gap in emissions between reported values from Annex I countries to the UNFCCC and global emissions derived from atmospheric measurements.

One piece of the missing emissions constituting the gap could perhaps be explained by improper accounting of emissions from (electronic) waste.

8.3. Global availability of information and geographical distribution of studies

Availability of data for different parts of the world is distributed very unequally. For Europe, the USA, China and Australia several bottom-up and top-down estimates were prepared. For China, reported data to the UNFCCC is however more incomplete, as it is not part of Annex I of the Kyoto Protocol.

For all countries of group 2 of Article 5 (Bahrain, India, Iran, Iraq, Kuwait, Oman, Pakistan, Qatar, Saudi Arabia, and the United Arab Emirates), for which a delayed phase-down schedule applies, limited to no information on HFC emissions is available. On the other hand, many non-Annex I countries voluntarily include some information on HFC consumption or emission estimates in their greenhouse gas assessments, for example Liberia.

8.4. Measurement station coverage

The addition of data from strategically placed measurement stations can significantly reduce uncertainty in top-down emission estimates and enable the quantification of emissions from further regions of the world, which are interesting with respect to the global emissions gap, such as Indonesia, Brazil, Iran and Saudi Arabia. HFC measurements from the AGAGE station Mt. Mugogo could be valuable for emission estimations for many African and Arab countries.

8.5. Key recommendations

Considering that important policy decisions depend on it, the basis of emission estimates should be robust. Emission estimates from different methods may vary considerably and gaps in global emissions cannot be fully explained. Therefore, more studies and further harmonization of the methods are valuable and needed.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2021.131208.

References

Arnold, T., Ivy, D.J., Harth, C.M., Vollmer, M.K., Mühle, J., Salameh, P.K., Paul Steele, L., Krummel, P.B., Wang, R.H.J., Young, D., Lunder, C.R., Hermansen, O., Rhee, T.S., Kim, J., Reimann, S., O'Doherty, S.J., Fraser, P.J., Simmonds, P.G., Prinn, R.G., Weiss, R.F., 2014. HFC-43-10mee atmospheric abundances and global emission estimates. Geophys. Res. Lett. 41 (6), 2228–2235. https://doi.org/10.1002/2013GL059143.

Ashford, P., 2016a. HFC Emissions Report for Bangladesh: Prepared under Contract to UNDP for CCAC. Anthesis-Caleb, p. 8. https://www.ccacoalition.org/en/resources/bangladesh-hfc-emissions-assessment.

Ashford, P., 2016b. HFC Emissions Report for Ghana: Prepared under Contract to UNDP for CCAC. Anthesis-Caleb, p. 7. https://www.ccacoalition.org/en/resources/ghana-hfc-emissions-assessment.

Ashford, P., 2016c. HFC Emissions Report for Moldova: Prepared under Contract to UNDP for US Government. Anthesis-Caleb, p. 7.

Ashford, P., 2016d. HFC Emissions Report for Nigeria: Prepared under Contract to UNDP for CCAC. Anthesis-Caleb, p. 7. https://www.ccacoalition.org/en/resources/nigeria-hfc-emissions-assessment.

Ashford, P., Baker, J.A., Clodic, D., Devotta, S., Godwin, D.S., Harnisch, J., Irving, W., Jeffs, M., Kuijpers, L., McCulloch, A., De Aguiar Peixoto, R., Uemura, S., Verdonik, D. P., Prepared by the National Greenhouse Gas Inventories Programme, 2006. Chapter 7: Emissions of Fluorinated Substitutes for Ozone Depleting Substances. IPCC Guidelines for National Greenhouse Gas Inventories, 3: Industrial Processes and Product Use. Institute for Global Environmental Strategies (IGES) on behalf of TheIntergovernmental Panel on Climate Change (IPCC), Hayama, Japan. https://www.ipcc-nggip.iges.or.jp/public/2006gl/index.html.

Azevedo, T.R. de, Costa Junior, C., Brandão Junior, A., Cremer, M.D.S., Piatto, M., Tsai, D.S., Barreto, P., Martins, H., Sales, M., Galuchi, T., Rodrigues, A., Morgado, R., Ferreira, A.L., Barcellos E Silva, F., Viscondi, G.d.F., Dos Santos, K.C., Cunha, K.B.d., Manetti, A., Coluna, I.M.E., Albuquerque, I.R.d., Junior, S.W., Leite, C., Kishinami, R., 2018. SEEG initiative estimates of Brazilian greenhouse gas emissions from 1970 to 2015. Sci. Data 5, 180045. https://doi.org/10.1038/sdata.2018.45.

Berends, A.G., Boutonnet, J.C., Rooij, C.G.D., Thompson, R.S., 1999. Toxicity of trifluoroacetate to aquatic organisms. Environ. Toxicol. Chem. 18 (5), 1053–1059. https://doi.org/10.1002/etc.5620180533.

Birmpili, T., 2018. Montreal protocol at 30: the governance structure, the evolution, and the Kigali amendment. C. R. Geosci. 350 (7), 425–431. https://doi.org/10.1016/j.crte.2018.09.002.

Brack, D., Climate and Clean Air Coalition (CCAC), United Nations Environment Programme (UNEP), United Nations Development Programme (UNDP), 2016. National Hydrocarbon (HFC) Inventories: A Summary of the Key Findings from the First Tranche of Studies. United Nations Environment Programme (UNEP), Nairobi, Kenya. https://www.ccacoalition.org/es/node/1714. (Accessed 11 July 2019).

Brunner, D., Arnold, T., Henne, S., Manning, A., Thompson, R.L., Maione, M., O'Doherty, S.J., Reimann, S., 2017. Comparison of four inverse modelling systems applied to the estimation of HFC-125, HFC-134a, and SF_6 emissions over Europe. Atmos. Chem. Phys. 17 (17), 10651–10674. https://doi.org/10.5194/acp-17-10651-2017.

Burkholder, J.B., Marshall, P., Bera, P.P., Francisco, J.S., Lee, T.J., 2020. Climate metrics for C1-C4 hydrofluorocarbons (HFCs). J. Phys. Chem. 124 (23), 4793–4800. https://doi.org/10.1021/acs.jpca.0c02679.

Chaturvedi, V., Sharma, M., 2015. Modelling long-term HFC emissions from India's residential air-conditioning sector: exploring implications of alternative refrigerants, best practices, and a sustainable lifestyle within an integrated assessment modelling framework. Clim. Pol. 16 (7), 877–893. https://doi.org/10.1080/14693062.2015.1052954.

Chaturvedi, V., Sharma, M., Chattopadhyay, S., Purohit, P., 2015. India's Long Term Hydrofluorocarbon Emissions: A Detailed Cross Sectoral Analysis within an Integrated Assessment Modelling Framework. Council on Energy, Environment and Water (CEEW); International Institute for Applied Systems Analysis (IIASA); Shakti Sustainable Energy Foundation, New Dehli, India. https://doi.org/10.13140/RG.2.1.1211.1524.

Cheewaphongphan, P., Chatani, S., Saigusa, N., 2019. Exploring gaps between bottom-up and top-down emission estimates based on uncertainties in multiple emission inventories: a case study on CH_4 emissions in China. Sustainability 11 (7), 2054. https://doi.org/10.3390/su11072054.

Climate and Clean Air Coalition (CCAC), United Nations Environment Programme (UNEP), 2016. Integrated Assessment of Short-Lived Climate Pollutants in Latin America and the Caribbean: Improving Air Quality while Contributing to Climate Change Mitigation. Summary for decision makers. United Nations Environment Programme (UNEP), Nairobi, Kenya. https://www.ccacoalition.org/es/node/1484.

Cousins, I.T., Ng, C.A., Wang, Z., Scheringer, M., 2019. Why is high persistence alone a major cause of concern? Environ. Sci. Proces. Impacts 21 (5), 781–792. https://doi.org/10.1039/c8em00515j.

Crippa, M., Oreggioni, G., Guizzardi, D., Muntean, M., Schaaf, E., Lo Vullo, E., Solazzo, E., Monforti-Ferrario, F., Olivier, J.G.J., Vignati, E., 2019. Fossil CO_2 and GHG Emissions of All World Countries: 2019 Report. Publications Office of the European Union, Luxembourg, Luxembourg, doi:10.2760/687800.

Earth Systems Research Laboratory (ESRL). HATS Flask Sampling Program: A brief history and current status. <https://www.esrl.noaa.gov/gmd/hats/flask/flasks.html> (retrieved 15.12.19).

Fang, X., Wu, J., Su, S., Han, J., Wu, Y., Shi, Y., Wan, D., Sun, X., Zhang, J., Hu, J., 2012. Estimates of major anthropogenic halocarbon emissions from China based on interspecies correlations. Atmos. Eviron. 62, 26–33. https://doi.org/10.1016/j.atmosenv.2012.08.010.

Fang, X., Velders, G.J.M., Ravishankara, A.R., Molina, M.J., Hu, J., Prinn, R.G., 2016. Hydrofluorocarbon (HFC) emissions in China: an inventory for 2005-2013 and projections to 2050. Environ. Sci. Technol. 50 (4), 2027–2034. https://doi.org/10.1021/acs.est.5b04376.

Energy and Environment Consultancy Joint Stock Company (VNEEC), 2017. HFC Inventory Socialist Republic of Viet Nam. The World Bank Group (World Bank), Washington, D.C., United States. https://www.ccacoalition.org/en/resources/vietnam-hfc-inventory.

Environmental Protection Agency Liberia (EPA Liberia), 2017. Ozone Depleting Substances Alternative Survey Report Liberia. Environmental Protection Agency Liberia (EPA Liberia), Monrovia, Liberia. https://epa.gov.lr/content/ozone-depleting-substances-alternative-survey-report-1.

Fang, X., Saito, T., Park, S., Li, S., Yokouchi, Y., Prinn, R.G., 2018. Performance of back-trajectory statistical methods and inverse modeling method in locating emission sources. ACS Earth Space Chem 2 (8), 843–851. https://doi.org/10.1021/acsearthspacechem.8b00062.

Fortems-Cheiney, A., Saunois, M., Pison, I., Chevallier, F., Bousquet, P., Cressot, C., Montzka, S.A., Fraser, P.J., Vollmer, M.K., Simmonds, P.G., Young, D., O'Doherty, S. J., Weiss, R.F., Artuso, F., Barletta, B., Blake, D.R., Li, S., Lunder, C., Miller, B.R., Park, S., Prinn, R., Saito, T., Steele, L.P., Yokouchi, Y., 2015. Increase in HFC-134a emissions in response to the success of the Montreal Protocol. J. Geophys. Res. Atmos. 120 (22) https://doi.org/10.1002/2015JD023741, 11,728-11,742.

Garg, A., Shukla, P.R., Kapshe, M., 2006. The sectoral trends of multigas emissions inventory of India. Atmos. Environ. 40 (24), 4608–4620. https://doi.org/10.1016/j.atmosenv.2006.03.045.

Ghandehari, M., Aghamohamadnia, M., Dobler, G., Karpf, A., Buckland, K., Qian, J., Koonin, S., 2017. Mapping refrigerant gases in the New York city skyline. Sci. Rep. 7 (1), 2735. https://doi.org/10.1038/s41598-017-02390-z.

Gobierno de México, 2018. Inventario nacional de emisiones de gases y compuestos de efecto invernadero 2017: Excel table. Gobierno de México. https://datos.gob.mx/busca/dataset/inventario-nacional-de-emisiones-de-gases-y-compuestos-de-efecto-invernadero-inegycei/resource/6b5b5c39-9185-4ef3-997c-81259cdc63c6?inner_span=True.

Graziosi, F., Arduini, J., Furlani, F., Giostra, U., Cristofanelli, P., Fang, X., Hermanssen, O., Lunder, C., Maenhout, G., O'Doherty, S.J., Reimann, S., Schmidbauer, N., Vollmer, M.K., Young, D., Maione, M., 2017. European emissions of the powerful greenhouse gases hydrofluorocarbons inferred from atmospheric measurements and their comparison with annual national reports to UNFCCC. Atmos. Eviron. 158, 85–97. https://doi.org/10.1016/j.atmosenv.2017.03.029.

Henne, S., Shallcross, D.E., Reimann, S., Xiao, P., Brunner, D., O'Doherty, S.J., Buchmann, B., 2012. Future emissions and atmospheric fate of HFC-1234yf from mobile air conditioners in Europe. Environ. Sci. Technol. 46 (3), 1650–1658. https://doi.org/10.1021/es2034608.

Henne, S., Vollmer, M.K., Steinbacher, M., Leuenberger, M., Meinhardt, F., Mohn, J., Emmenegger, L., Brunner, D., Reimann, S., 2020. Top-down Support of Swiss non-CO_2 Greenhouse Gas Emissions Reporting to UNFCCC. EGU General Assembly 2020. https://doi.org/10.5194/egusphere-egu2020-17176.

Hodnebrog, Ø., Etminan, M., Fuglestvedt, J.S., Marston, G., Myhre, G., Nielsen, C.J., Shine, K.P., Wallington, T.J., 2013. Global warming potentials and radiative efficiencies of halocarbons and related compounds: a comprehensive review. Rev. Geophys. 51 (2), 300–378. https://doi.org/10.1002/rog.20013.

Hu, L., Montzka, S.A., Miller, J.B., Andrews, A.E., Lehman, S.J., Miller, B.R., Thoning, K., Sweeney, C., Chen, H., Godwin, D.S., Masarie, K., Bruhwiler, L., Fischer, M.L., Biraud, S.C., Torn, M.S., Mountain, M., Nehrkorn, T., Eluszkiewicz, J., Miller, S., Draxler, R.R., Stein, A.F., Hall, B.D., Elkins, J.W., Tans, P.P., 2015. U.S. emissions of HFC-134a derived for 2008-2012 from an extensive flask-air sampling network. J. Geophys. Res. Atmos. 120 (2), 801–825. https://doi.org/10.1002/2014JD022617.

Hu, L., Montzka, S.A., Lehman, S.J., Godwin, D.S., Miller, B.R., Andrews, A.E., Thoning, K., Miller, J.B., Sweeney, C., Siso, C., Elkins, J.W., Hall, B.D., Mondeel, D. J., Nance, D., Nehrkorn, T., Mountain, M., Fischer, M.L., Biraud, S.C., Chen, H., Tans, P.P., 2017. Considerable contribution of the Montreal Protocol to declining greenhouse gas emissions from the United States. Geophys. Res. Lett. 44 (15), 8075–8083. https://doi.org/10.1002/2017GL074388.

Hurwitz, M.M., Fleming, E.L., Newman, P.A., Li, F., Mlawer, E., Cady-Pereira, K., Bailey, R., 2015. Ozone depletion by hydrofluorocarbons. Geophys. Res. Lett. 42 (20), 8686–8692. https://doi.org/10.1002/2015GL065856.

Jung, K., Ro, D., Park, Y.-K., 2020. Estimation, and framework proposal of greenhouse gas emissions of fluorinated substitutes for ozone-depleting substances by application area in the Republic of Korea. Sustainability 12 (16), 6355. https://doi.org/10.3390/su12166355.

Keller, C.A., Hill, M., Vollmer, M.K., Henne, S., Brunner, D., Reimann, S., O'Doherty, S.J., Arduini, J., Maione, M., Ferenczi, Z., Haszpra, L., Manning, A.J., Peter, T., 2012. European emissions of halogenated greenhouse gases inferred from atmospheric measurements. Environ. Sci. Technol. 46 (1), 217–225. https://doi.org/10.1021/es202453j.

Kim, J., Li, S., Kim, K.-R., Stohl, A., Mühle, J., Kim, S.-K., Park, M.-K., Kang, D.-J., Lee, G., Harth, C.M., Salameh, P.K., Weiss, R.F., 2010. Regional atmospheric emissions determined from measurements at Jeju Island, Korea: halogenated compounds from China. Geophys. Res. Lett. 37 (12) n/a-n/a. doi:10.1029/2010GL043263.

Kuyper, B., Say, D., Labuschagne, C., Lesch, T., Joubert, W.R., Martin, D., Young, D., Khan, M.A.H., Rigby, M., Ganesan, A.L., Lunt, M.F., O'Dowd, C., Manning, A.J., O'Doherty, S.J., Davies-Coleman, M.T., Shallcross, D.E., 2019. Atmospheric HCFC-22, HFC-125, and HFC-152a at Cape point, South Africa. Environ. Sci. Technol. 53 (15), 8967–8975. https://doi.org/10.1021/acs.est.9b01612.

LaDou, J., Lovegrove, S., 2008. Export of electronics equipment waste. Int. J. Occup. Environ. Health 14 (1), 1–10. https://doi.org/10.1179/oeh.2008.14.1.1.

Li, S., Kim, J., Kim, K.-R., Mühle, J., Kim, S.-K., Park, M.-K., Stohl, A., Kang, D.-J., Arnold, T., Harth, C.M., Salameh, P.K., Weiss, R.F., 2011. Emissions of halogenated compounds in east Asia determined from measurements at Jeju island, Korea. Environ. Sci. Technol. 45 (13), 5668–5675. https://doi.org/10.1021/es104124k.

Lindley, A., McCulloch, A., Vink, T., 2019. Contribution of hydrofluorocarbons (HFCs) and hydrofluoro-olefins (HFOs) atmospheric breakdown products to acidification ("Acid rain") in the EU at present and in the future. Open J. Air Pollut. 81–95. https://doi.org/10.4236/ojap.2019.84004, 08 (04).

Liu, L., Dou, Y., Yao, B., Bie, P., Wang, L., Peng, M., Hu, J., 2019. Historical and projected HFC-410A emission from room air conditioning sector in China. Atmos. Eviron. 212, 194–200. https://doi.org/10.1016/j.atmosenv.2019.05.022.

Luecken, D.J., L Waterland, R., Papasavva, S., Taddonio, K.N., Hutzell, W.T., Rugh, J.P., Andersen, S.O., 2010. Ozone and TFA impacts in North America from degradation of 2,3,3,3-Tetrafluoropropene (HFO-1234yf), a potential greenhouse gas replacement. Environ. Sci. Technol. 44 (1), 343–348. https://doi.org/10.1021/es902481f.

Lunt, M.F., Rigby, M., Ganesan, A.L., Manning, A.J., Prinn, R.G., O'Doherty, S.J., Mühle, J., Harth, C.M., Salameh, P.K., Arnold, T., Weiss, R.F., Saito, T., Yokouchi, Y., Krummel, P.B., Steele, L.P., Fraser, P.J., Li, S., Park, S., Reimann, S., Vollmer, M.K., Lunder, C., Hermansen, O., Schmidbauer, N., Maione, M., Arduini, J., Young, D., Simmonds, P.G., 2015. Reconciling reported and unreported HFC emissions with atmospheric observations. Proc. Natl. Acad. Sci. U.S.A. 112 (19), 5927–5931. https://doi.org/10.1073/pnas.1420247112.

Miller, B.R., Weiss, R.F., Salameh, P.K., Tanhua, T., Greally, B.R., Mühle, J., Simmonds, P.G., 2008. Medusa: a sample preconcentration and GC/MS detector system for in situ measurements of atmospheric trace halocarbons, hydrocarbons,

and sulfur compounds. Anal. Chem. 80 (5), 1536–1545. https://doi.org/10.1021/ac702084k.
Millet, D.B., Atlas, E.L., Blake, D.R., Blake, N.J., Diskin, G.S., Holloway, J.S., Hudman, R. C., Meinardi, S., Ryerson, T.B., Sachse, G.W., 2009. Halocarbon emissions from the United States and Mexico and their global warming potential. Environ. Sci. Technol. 43 (4), 1055–1060. https://doi.org/10.1021/es802146j.
Ministério Da Ciência, Tecnologia E Inovação, 2017. ESTIMATIVAS ANUAIS DE EMISSÕES DE GASES DE EFEITO ESTUFA NO BRASIL: 4a Edição. Ministry of Science, Technology and Innovation,Government of Brasil, Brasília, Brazil. https://antigo.mctic.gov.br/mctic/opencms/ciencia/SEPED/clima/Comunicacao_Nacional/Estimativas_Anuais.html.
Ministry of Environment Chile, 2014. Survey of the Hydrofluorocarbon (HFC) Market in Chile. Final Report. Ministry of Environment, Government of Chile, Santiago, Chile. https://www.ccacoalition.org/en/resources/chile-hfc-inventory.
Montzka, S.A., McFarland, M., Andersen, S.O., Miller, B.R., Fahey, D.W., Hall, B.D., Hu, L., Siso, C., Elkins, J.W., 2015. Recent trends in global emissions of hydrochlorofluorocarbons and hydrofluorocarbons: reflecting on the 2007 adjustments to the Montreal Protocol. J. Phys. Chem. 119 (19), 4439–4449. https://doi.org/10.1021/jp5097376.
Montzka, S.A., Velders, G.J.M., Krummel, P.B., Mühle, J., Orkin, V.L., Park, S., Shah, N., Walter-Terrinoni, H., 2018. Hydrofluorocarbons: Chapter 2. Scientific Assessment of Ozone Depletion: 2018. Global Ozone Research and Monitoring Project-Report No. 58, Geneva, Switzerland.
MIT, National Aeronautics and Space Administration USA (NASA). Advanced Global Atmospheric Gases Experiment (AGAGE) Website. <https://agage.mit.edu/> (retrieved 13.12.19).
National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA), 2010-2020. ENEA Station for Climate Observation Roberto Sarao: Activities. <http://www.lampedusa.enea.it/attivita/index.php?lang=en> (retrieved 24.12.19).
Netherlands Environmental Assessment Agency (PBL), European Commission Joint Research Center (EC-JRC), 2017. EDGAR (Emission Database for Global Atmospheric Research). European Commission Joint Research Center (EC-JRC), Brussels, Belgium release version 4.2 FT2010, 2011. https://edgar.jrc.ec.europa.eu/. (Accessed 15 December 2019).
Nisbet, E., Weiss, R., 2010. Atmospheric science. Top-down versus bottom-up. Science 328 (5983), 1241–1243. https://doi.org/10.1126/science.1189936.
O'Doherty, S.J., Cunnold, D.M., Miller, B.R., Mühle, J., McCulloch, A., Simmonds, P.G., Manning, A.J., Reimann, S., Vollmer, M.K., Greally, B.R., Prinn, R.G., Fraser, P.J., Steele, L.P., Krummel, P.B., Dunse, B.L., Porter, L.W., Lunder, C.R., Schmidbauer, N., Hermansen, O., Salameh, P.K., Harth, C.M., Wang, R.H.J., Weiss, R.F., 2009. Global and regional emissions of HFC-125 (CHF 2 CF 3) from in situ and air archive atmospheric observations at AGAGE and SOGE observatories. J. Geophys. Res. 114 (D23), 125. https://doi.org/10.1029/2009JD012184.
Omotosho, D.B., 2015. HFC Inventory Nigeria: 2008-2014. United Nations Development Program (UNDP), Climate and Clean Air Coalition (CCAC), Paris, France. https://www.ccacoalition.org/en/resources/nigeria-hfc-inventory.
Papasavva, S., Luecken, D.J., Waterland, R.L., Taddonio, K.N., Andersen, S.O., 2009. Estimated 2017 refrigerant emissions of 2,3,3,3-tetrafluoropropene (HFC-1234yf) in the United States resulting from automobile air conditioning. Environ. Sci. Technol. 43 (24), 9252–9259. https://doi.org/10.1021/es902124u.
Pasek, A.D., 2014. HFC Inventory Indonesia (2010-2012). Climate and Clean Air Coalition (CCAC). United Nations Development Programme (UNDP), Climate and Clean Air Coalition (CCAC), Paris, France. https://ccacoalition.org/en/resource-library.
National Ozone Unit of Colombia (UTO), United Nations Development Programme (UNDP), 2014. Study of the Hydrofluorocarbons (HFCs) Market in Colombia. Final report. National Ozone Unit of Colombia (UTO), Bogotá, Colombia. https://www.ccacoalition.org/en/resources/colombia-hfc-inventory.
Pickard, H.M., Criscitiello, A.S., Persaud, D., Spencer, C., Muir, D.C.G., Lehnherr, I., Sharp, M.J., Silva, A.O. de, Young, C.J., 2020. Ice core record of persistent short-chain fluorinated alkyl acids: evidence of the impact from global environmental regulations. Geophys. Res. Lett. 47 (10), 65. https://doi.org/10.1029/2020GL087535.
Prinn, R.G., Weiss, R.F., Fraser, P.J., Simmonds, P.G., Cunnold, D.M., Alyea, F.N., O'Doherty, S., Salameh, P., Miller, B.R., Huang, J., Wang, R.H.J., Hartley, D.E., Harth, C., Steele, L.P., Sturrock, G., Midgley, P.M., McCulloch, A., 2000. A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE. J. Geophys. Res. 105 (D14), 17751–17792. https://doi.org/10.1029/2000JD900141.
Pu, J., Xu, H., Yao, B., Yu, Y., Jiang, Y., Ma, Q., Chen, L., 2020. Estimate of hydrofluorocarbon emissions for 2012–16 in the Yangtze River Delta, China. Adv. Atmos. Sci. 37 (6), 576–585. https://doi.org/10.1007/s00376-020-9242-3.
Purohit, P., Höglund-Isaksson, L., 2017. Global emissions of fluorinated greenhouse gases 2005–2050 with abatement potentials and costs. Atmos. Chem. Phys. 17 (4), 2795–2816. https://doi.org/10.5194/acp-17-2795-2017.
29.10.19 Purohit, P., Höglund-Isaksson, L., Wagner, F., 2018. Impacts of the Kigali amendment to phase-down hydrofluorocarbons (HFCs) in Asia. IIASA report. International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria. http://pure.iiasa.ac.at/id/eprint/15274/.
Purohit, P., Höglund-Isaksson, L., Dulac, J., Shah, N., Wei, M., Rafaj, P., Schöpp, W., 2020. Electricity savings and greenhouse gas emission reductions from global phase-down of hydrofluorocarbons. Atmos. Chem. Phys. 20 (19), 11305–11327. https://doi.org/10.5194/acp-20-11305-2020.
Ramanathan, V., 1975. Greenhouse effect due to chlorofluorocarbons: climatic implications. Science 190 (4209), 50–52. https://doi.org/10.1126/science.190.4209.50.
Ravishankara, A.R., Turnipseed, A.A., Jensen, N.R., Barone, S., Mills, M., Howard, C.J., Solomon, S., 1994. Do hydrofluorocarbons destroy stratospheric ozone? Science 263 (5143), 71–75. https://doi.org/10.1126/science.263.5143.71.
Reazuddin, M., 2014. HFC Inventory Bangladesh (2011-2013). United Nations Development Program (UNDP), Climate and Clean Air Coalition (CCAC), Paris, France. https://www.ccacoalition.org/en/resources/bangladesh-hfc-emissions-assessment.
Say, D., Manning, A.J., O'Doherty, S.J., Rigby, M., Young, D., Grant, A., 2016. Re-evaluation of the UK's HFC-134a emissions inventory based on atmospheric observations. Environ. Sci. Technol. 50 (20), 11129–11136. https://doi.org/10.1021/acs.est.6b03630.
UNEP Refrigeration, Air Conditioning and Heat Pumps Technical Options Committee (RTOC), 2019. 2018 Assessment Report of the Refrigeration UNEP Refrigeration, Air Conditioning and Heat Pumps Technical Options Committee. United Nations Environment Program (UNEP), Nairobi, Kenya. https://ozone.unep.org/sites/default/files/2019-04/RTOC-assessment-report-2018_0.pdf.
Say, D., Ganesan, A.L., Lunt, M.F., Rigby, M., O'Doherty, S.J., Harth, C., Manning, A.J., Krummel, P.B., Bauguitte, S., 2019. Emissions of halocarbons from India inferred through atmospheric measurements. Atmos. Chem. Phys. 19 (15), 9865–9885. https://doi.org/10.5194/acp-19-9865-2019.
Schoenenberger, F., Henne, S., Hill, M., Vollmer, M.K., Kouvarakis, G., Mihalopoulos, N., O'Doherty, S.J., Maione, M., Emmenegger, L., Peter, T., Reimann, S., 2018. Abundance and sources of atmospheric halocarbons in the Eastern Mediterranean. Atmos. Chem. Phys. 18 (6), 4069–4092. https://doi.org/10.5194/acp-18-4069-2018.
Ozone Secretariat, 2018. In: Handbook for the Montreal Protocol on Substances that Deplete the Ozone Layer, twelfth ed. United Nations Environment Program (UNEP), Nairobi, Kenya. ISBN: 978-9966-076-33-5.
Sharma, S.K., Choudhury, A., Sarkar, P., Biswas, S., Singh, A., Dadhich, P.K., Singh, A.K., Majumdar, S., Bhatia, A., Mohini, M., Kumar, R., Jha, C.S., Murthy, M.S.R., Ravindranath, N.H., Bhattacharya, J.K., Karthik, M., Bhattacharya, S., Chauhan, R., 2011. Greenhouse gas inventory estimates for India. Curr. Sci. 101 (3). https://www.jstor.org/stable/24078519.
Simmonds, P.G., Derwent, R.G., Manning, A.J., McCulloch, A., O'Doherty, S.J., 2015. USA emissions estimates of CH3CHF2, CH2FCF3, CH3CF3 and CH2F2 based on in situ observations at Mace Head. Atmos. Eviron. 104, 27–38. https://doi.org/10.1016/j.atmosenv.2015.01.010.
Simmonds, P.G., Rigby, M., Manning, A.J., Lunt, M.F., O'Doherty, S.J., McCulloch, A., Fraser, P.J., Henne, S., Vollmer, M.K., Mühle, J., Weiss, R.F., Salameh, P.K., Young, D., Reimann, S., Wenger, A., Arnold, T., Harth, C.M., Krummel, P.B., Steele, L.P., Dunse, B.L., Miller, B.R., Lunder, C.R., Hermansen, O., Schmidbauer, N., Saito, T., Yokouchi, Y., Park, S., Li, S., Yao, B., Zhou, L.X., Arduini, J., Maione, M., Wang, R.H.J., Ivy, D., Prinn, R.G., 2016. Global and regional emissions estimates of 1,1-difluoroethane (HFC-152a, CH_3CHF_2) from in situ and air archive observations. Atmos. Chem. Phys. 16 (1), 365–382. https://doi.org/10.5194/acp-16-365-2016.
Simmonds, P.G., Rigby, M., McCulloch, A., O'Doherty, S.J., Young, D., Mühle, J., Krummel, P.B., Steele, P., Fraser, P.J., Manning, A.J., Weiss, R.F., Salameh, P.K., Harth, C.M., Wang, R.H.J., Prinn, R.G., 2017. Changing trends and emissions of hydrochlorofluorocarbons (HCFCs) and their hydrofluorocarbon (HFCs) replacements. Atmos. Chem. Phys. 17 (7), 4641–4655. https://doi.org/10.5194/acp-17-4641-2017.
Simmonds, P.G., Palmer, P.I., Rigby, M., McCulloch, A., O'Doherty, S., Manning, A.J., 2020. Tracers for Evaluating Computational Models of Atmospheric Transport and Oxidation at Regional to Global Scales. Atmospheric Environment, p. 118074. https://doi.org/10.1016/j.atmosenv.2020.118074.
Solomon, K.R., Velders, G.J.M., Wilson, S.R., Madronich, S., Longstreth, J., Aucamp, P.J., Bornman, J.F., 2016. Sources, fates, toxicity, and risks of trifluoroacetic acid and its salts: relevance to substances regulated under the Montreal and Kyoto Protocols. J. Toxicol. Environ. Health B Crit. Rev. 19 (7), 289–304. https://doi.org/10.1080/10937404.2016.1175981.
Stemmler, K., Folini, D., Ubl, S., Vollmer, M.K., Reimann, S., O'Doherty, S.J., Greally, B. R., Simmonds, P.G., Manning, A.J., 2007. European emissions of HFC-365mfc, a chlorine-free substitute for the foam blowing agents HCFC-141b and CFC-11. Environ. Sci. Technol. 41 (4), 1145–1151. https://doi.org/10.1021/es061298h.
Sthiannopkao, S., Wong, M.H., 2013. Handling e-waste in developed and developing countries: initiatives, practices, and consequences. Sci. Total Environ. 463–464, 1147–1153. https://doi.org/10.1016/j.scitotenv.2012.06.088.
Stohl, A., Seibert, P., Arduini, J., Eckhardt, S., Fraser, P., Greally, B.R., Lunder, C., Maione, M., Mühle, J., O'Doherty, S.J., Prinn, R.G., Reimann, S., Saito, T., Schmidbauer, N., Simmonds, P.G., Vollmer, M.K., Weiss, R.F., Yokouchi, Y., 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. https://doi.org/10.5194/acp-9-1597-2009.
Stohl, A., Kim, J., Li, S., O'Doherty, S.J., Mühle, J., Salameh, P.K., Saito, T., Vollmer, M. K., Wan, D., Weiss, R.F., Yao, B., Yokouchi, Y., Zhou, L.X., 2010. Hydrochlorofluorocarbon and hydrofluorocarbon emissions in East Asia determined by inverse modeling. Atmos. Chem. Phys. 10 (8), 3545–3560. https://doi.org/10.5194/acp-10-3545-2010.
Strahan, S.E., Douglass, A.R., 2018. Decline in antarctic ozone depletion and lower stratospheric chlorine determined from aura microwave limb sounder observations. Geophys. Res. Lett. 45 (1), 382–390. https://doi.org/10.1002/2017GL074830.
Strong, K., Godin, P.J., Le Bris, K., Johnson, H., Piunno, R., Cabaj, A., MacDougall, C., Xu, L.H., 2018. Infrared absorption cross-sections of fluorinated molecules for

improved global warming potential calculations. Am. Geophys. Union A33L–3349. Fall Meeting 2018, abstract #A33L-3349. AGU Fall Meeting Abstracts 2018.
Su, S., Fang, X., Li, L., Wu, J., Zhang, J., Xu, W., Hu, J., 2015. HFC-134a emissions from mobile air conditioning in China from 1995 to 2030. Atmos. Eviron. 102, 122–129. https://doi.org/10.1016/j.atmosenv.2014.11.057.
Tohka, A., 2005. The GAINS model for greenhouse gases – version 1.0: HFC, PFC and SF6. IIASA Interim Report IR-05-56, 67 pp.
Tovar, C.M., Blanco, M.B., Barnes, I., Wiesen, P., Teruel, M.A., 2014. Gas-phase reactivity study of a series of hydrofluoroolefins (HFOs) toward OH radicals and Cl atoms at atmospheric pressure and 298 K. Atmos. Environ. 88 (8), 107–114. https://doi.org/10.1016/j.atmosenv.2014.01.060.
United Nations Framework Convention on Climate Change (UNFCCC), 2020a. Greenhousegas Inventory Data: Flexible Queries Non Annex I Countries, 12.12.19-04.01.21. https://di.unfccc.int/flex_non_annex1.
United Nations Framework Convention on Climate Change (UNFCCC), 2020b. Greenhousegas Inventory Data: Flexible Queries Annex I Countries, 12.12.19-04.01.21. https://di.unfccc.int/flex_annex1.
United Nations Industrial Development Organization (UNIDO), 2017. HFC Inventory Report South Africa. United Nations Industrial Development Organization (UNIDO), Vienna, Austria. https://www.ccacoalition.org/en/resources/south-africa-hfc-inventory.
United Nations Industrial Development Organization (UNIDO), 2018. HFC Inventory of Jordan. Final Report. United Nations Industrial Development Organization (UNIDO), Vienna, Austria. https://www.ccacoalition.org/en/resources/jordan-hfc-inventory.
Velders, G.J.M., Andersen, S.O., Daniel, J.S., Fahey, D.W., McFarland, M., 2007. The importance of the Montreal Protocol in protecting climate. Proc. Natl. Acad. Sci. U.S.A. 104 (12), 4814–4819. https://doi.org/10.1073/pnas.0610328104.
Velders, G.J.M., Fahey, D.W., Daniel, J.S., McFarland, M., Andersen, S.O., 2009. The large contribution of projected HFC emissions to future climate forcing. Proc. Natl. Acad. Sci. U.S.A. 106 (27), 10949–10954. https://doi.org/10.1073/pnas.0902817106.
Velders, G.J.M., Ravishankara, A.R., Miller, M.K., Molina, M.J., Alcamo, J., Daniel, J.S., Fahey, D.W., Montzka, S.A., Reimann, S., 2012. Climate change. Preserving Montreal Protocol climate benefits by limiting HFCs. Science 335 (6071), 922–923. https://doi.org/10.1126/science.1216414.
Velders, G.J.M., Solomon, S., Daniel, J.S., 2014. Growth of climate change commitments from HFC banks and emissions. Atmos. Chem. Phys. 14 (9), 4563–4572. https://doi.org/10.5194/acp-14-4563-2014.
Velders, G.J.M., Fahey, D.W., Daniel, J.S., Andersen, S.O., McFarland, M., 2015. Future atmospheric abundances and climate forcings from scenarios of global and regional hydrofluorocarbon (HFC) emissions. Atmos. Eviron. 123, 200–209. https://doi.org/10.1016/j.atmosenv.2015.10.071.
Dunse, B.L., Derek, N., Fraser, P.J., Krummel, P.B., Steele, L.P., 2018. Australian and Global HFC, PFC, Sulfur Hexafluoride Nitrogen Trifluoride and Sulfuryl Fluoride Emissions: Report Prepared for Australian Government Department of the Environment and Energy. CSIRO Oceans and Atmosphere, Aspendale, Australia.
Vollmer, M.K., Miller, B.R., Rigby, M., Reimann, S., Mühle, J., Krummel, P.B., O'Doherty, S.J., Kim, J., Rhee, T.S., Weiss, R.F., Fraser, P.J., Simmonds, P.G., Salameh, P.K., Harth, C.M., Wang, R.H.J., Steele, L.P., Young, D., Lunder, C.R., Hermansen, O., Ivy, D., Arnold, T., Schmidbauer, N., Kim, K.-R., Greally, B.R., Hill, M., Leist, M., Wenger, A., Prinn, R.G., 2011. Atmospheric histories and global emissions of the anthropogenic hydrofluorocarbons HFC-365mfc, HFC-245fa, HFC-227ea, and HFC-236fa. J. Geophys. Res. 116 (D8), 701. https://doi.org/10.1029/2010JD015309.
Vollmer, M.K., Reimann, S., Hill, M., Brunner, D., 2015. First observations of the fourth generation synthetic halocarbons HFC-1234yf, HFC-1234ze(E), and HCFC-1233zd (E) in the atmosphere. Environ. Sci. Technol. 49 (5), 2703–2708. https://doi.org/10.1021/es505123x.
Wang, Z., Wang, Y., Li, J., Henne, S., Zhang, B., Hu, J., Zhang, J., 2018. Impacts of the degradation of 2,3,3,3-tetrafluoropropene into trifluoroacetic acid from its application in automobile air conditioners in China, the United States, and Europe. Environ. Sci. Technol. 52 (5), 2819–2826. https://doi.org/10.1021/acs.est.7b05960.
Wofsy, S.C., 2011. HIAPER Pole-to-Pole Observations (HIPPO): fine-grained, global-scale measurements of climatically important atmospheric gases and aerosols. Phil. Trans. A, Math. Phys. Eng. Sci. 369 (1943), 2073–2086. https://doi.org/10.1098/rsta.2010.0313.
Technology and Economic Assessment Panel (TEAP), 2019. 2018 Assessment Report of the Technology and Economic Assessment Panel. United Nations Environment Program (UNEP), Nairobi, Kenya. https://ozone.unep.org/science/assessment/teap.
World Bank Open Data: Total Greenhouse Gas Emissions; GDP Per Capita, 2019. The World Bank Group (World Bank), Washington, D.C., United States of America. https://data.worldbank.org/. (Accessed 14 December 2019).
Xiang, B., Patra, P.K., Montzka, S.A., Miller, S.M., Elkins, J.W., Moore, F.L., Atlas, E.L., Miller, B.R., Weiss, R.F., Prinn, R.G., Wofsy, S.C., 2014. Global emissions of refrigerants HCFC-22 and HFC-134a: unforeseen seasonal contributions. Proc. Natl. Acad. Sci. U.S.A. 111 (49), 17379–17384. https://doi.org/10.1073/pnas.1417372111.
Xu, Y., Zaelke, D., Velders, G.J.M., Ramanathan, V., 2013. The role of HFCs in mitigating 21st century climate change. Atmos. Chem. Phys. 13 (12), 6083–6089. https://doi.org/10.5194/acp-13-6083-2013.
Yao, B., Vollmer, M.K., Zhou, L.X., Henne, S., Reimann, S., Li, P.C., Wenger, A., Hill, M., 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. https://doi.org/10.5194/acp-12-10181-2012.
Yao, B., Fang, X., Vollmer, M.K., Reimann, S., Chen, L., Fang, S., Prinn, R.G., 2019. China's hydrofluorocarbon emissions for 2011–2017 inferred from atmospheric measurements. Environ. Sci. Technol. Lett. 6 (8), 479–486. https://doi.org/10.1021/acs.estlett.9b00319.
Yokouchi, Y., Taguchi, S., Saito, T., Tohjima, Y., Tanimoto, H., Mukai, H., 2006. High frequency measurements of HFCs at a remote site in east Asia and their implications for Chinese emissions. Geophys. Res. Lett. 33 (21), D15S03. https://doi.org/10.1029/2006GL026403.
Zeng, L., Dang, J., Guo, H., Lyu, X., Simpson, I.J., Meinardi, S., Wang, Y., Zhang, L., Blake, D.R., 2020. Long-term temporal variations and source changes of halocarbons in the Greater Pearl River Delta region, China. Atmos. Environ. 234, 117550. https://doi.org/10.1016/j.atmosenv.2020.117550.
Zhang, J., Wang, C., 2014. China's hydrofluorocarbon challenge. Nat. Clim. Change 4 (11), 943–945. https://doi.org/10.1038/nclimate2377.
Zhao, L., Zeng, W., Yuan, Z., 2015. Reduction of potential greenhouse gas emissions of room air-conditioner refrigerants: a life cycle carbon footprint analysis. J. Clean. Prod. 100, 262–268. https://doi.org/10.1016/j.jclepro.2015.03.063.