Author responses to peer review of manuscripts '*Emissions of CFCs, HCFCs and HFCs from India*' and '*Atmospheric observations and emissions estimates of ozone-depleting chlorocarbons from India*'

Daniel Say on behalf of all co-authors

We thank the anonymous reviewers for their useful feedback on our manuscripts. Please find below responses to their constructive comments. The merger has resulted in a number of changes to the manuscript – these are shown in the marked-up version where possible. Major changes are listed below.

AUTHOR NOTES:

1. Please note that the manuscripts **acp-2018-1146** and **acp-2018-1287** have been merged upon consideration of the comments made by the reviewers of acp-2018-1287. Here follow responses to reviews from both manuscripts, in the order they were submitted. Several of the comments made with respect to **acp-2018-1287** are no longer relevant as a result of the merger – these have been indicated as such.
2. During the review of **acp-2018-1146**, the measurements in Fig. 2 were found to be presented in an incorrect order. This Figure has been replaced by a corrected version - the measurement order is now consistent with those shown in Fig. 5.
3. As a result of the time that has elapsed since submission, the global warming potentials originally quoted are now out of date. These have been replaced in the merged manuscript by those presented in the 2018 Scientific Assessment on Ozone Depletion. Emissions reported in Tg CO2e have been updated accordingly. In all cases the differences are small and do not affect the outcome of the manuscript.
4. Our estimate of India's dichloromethane and chloroform emissions were quoted in error (**acp-2018-1287**). We had mistakenly quoted the NCI total rather than the India total. The correct estimates are 96.5 (77.8 - 115.6) Gg yr-1 (India) and 32.2 (28.3 – 37.1) Gg yr-1 (NCI) - the manuscript has been updated accordingly and the authors apologise for the oversight.
5. In addition to the correlations presented in both **acp-2018-1146** and **acp-2018-1287**, we add the scatter plot of HFC-23 versus chloroform to add further information on the possible sources of chloroform.
6. We remove our previous assertion that the majority of chloroform emissions are from anthropogenic sources. While the correlation with DCM is significant, we do not believe it is strong enough to justify our previous assertion. Since we now believe there to be biogenic emissions of chloroform, we do not scale NCI chloroform emissions to a national total.
7. Our updated paragraph on the production of CTC as a bi-product of chloromethane manufacture now includes discussion alluding to the fact that the difference between our top-down estimate, and the theoretical mass of CTC produced by these facilities, could be due to consumption of CTC in industries such as the production of divinyl acid chloride (P15 L6-8).

Responses to Review 1, acp-2018-1146

Reviewer: Major issues: In the title it could be mentioned that this is based on measurements.
Response: We agree that the title of the manuscript should reflect the fact that the emission estimates are based on atmospheric measurements. Hence, the merged title now reads *'Emissions of halocarbons from India inferred through atmospheric observations'*.

Reviewer: P1. L 3. Existing atmospheric measurement networks.
Response: Suggestion accepted and added to manuscript (P1 L3).

Reviewer: L 6. Use km instead of miles.
Response: Suggestion accepted and added to manuscript (P1 L6).

Reviewer: L 12. Our total CFCs.
Response: Based on the advice of the reviewer, we add the slightly modified '*Our combined CFC estimates*', to ensure that readers appreciate that the proceeding estimates are the combined emissions of CFC-11, CFC-12 and CFC-113 (P1 L13).

Reviewer: L14. I would delete the second part of the sentence (starting from, suggesting. . .), as this does not mean anything.
Response: Suggestion accepted – *'suggesting that India used a range of HCFC and HFC refrigerants in 2016'* removed from the manuscript.

Reviewer: P2 L2. Wallington seems to be a pretty inappropriate reference for this. Either one of the recent ozone assessments could be cited or Molina and Rowland.
Response: We accept that a more appropriate reference(s) would be fitting. Hence, we replace Wallington *et al.,* 1994 with Molina and Rowland 1974 and the 2018 Scientific Assessment of Ozone Depletion: Chapter 1 (Engel *et al.,* 2019) on P2 L9.

Reviewer: L 6. Derwent, Velders, inappropriate: one of the recent ozone assessments would be much better.
Response: We replace Derwent et al., and Velders et al., with the 2018 Scientific Assessment of Ozone Depletion: Chapter 1 (Engel *et al.,* 2014) on P2 L11.

Reviewer: L 9. ODSs.
Response: Suggestion accepted and manuscript updated accordingly (P2 L11).

Reviewer: L 10. Wrong! Emissions have been reduced in the last decade. But they are to some degree re-increasing.
Response: We are not entirely clear on the referee comment here, which does say they are re-increasing. Our statement in the manuscript states "*While the emissions of many ODSs are declining, broadly in line with expectations, the emissions of some species, CFC-11 in particular, have not reduced in the last decade, and are now increasing (Montzka et al., 2018)*." Montzka *et al.,* 2018 reported relatively constant global emissions of CFC-11 from 2002 – 2012 (54 ± 3 Gg yr^{-1}), followed by an increase of 13.5 ± 5 Gg yr^{-1} to 67 ± 3 Gg yr^{-1} (2014 - 2016). Hence, we do not make any changes to the manuscript with regards to this comment.

Reviewer: L 11. Be precise: Montzka as Southeast Asia.
Response: Rigby et al., 2019 (Nature) is now in press, and suggests the increase is due to emissions from China. We update the manuscript to state this, P2 L16.

Reviewer: L 16. Article 5 countries (developing countries) . . . remark, nobody outside the Montreal protocol knows that.
Response: We do not add this definition here because we define developing (Article 5) countries earlier on P2 L12.

Reviewer: L 17. . .currently still permitted. . .
Response: We replace 'currently' with 'still' on P2 L24.

Reviewer: P 6 L 18 not all emissions are on-going so I suggest: . . .of these gases could be ongoing. . .
Response: Since we only have evidence of continued emissions for CFC-11, we change *'emissions of these gases are ongoing'* to *'emissions of these gases could be ongoing'* on P7 L23.

Reviewer: L 20 make a reference to section 2.6, where the model is explained.
Response: Added reference to section 2.6 on P7 L25 – *'(an extension of the work by Rigby et al., 2014, see section 2.6)'*.

Reviewer: P 8 L 1. It is strange here obviously new lifetimes are used but in the table 1 still the outdated lifetimes of Myhre are used. The lifetimes from the SPARC report should be used in the table. The GWPs could still be from Myhre.
Response: All atmospheric lifetimes are now taken from the 2018 Scientific Assessment on Ozone Depletion (Engel et al., 2019).

Reviewer: L 22-26. This is said again behind. Delete it at one place.
Response: We remove *'due to the prevailing westerly winds that bring well-mixed oceanic air to the Indian subcontinent during these months. Back trajectory analysis confirmed that these samples had not interacted with any other significant land mass in the 30 days prior to collection.'* in order to prevent repetition of statements.

We also delete *'Except for HFC-134a, the measurements derived from these samples exhibited very little variation, and the mole fractions were amongst the lowest observed during the campaign, which was consistent with the oceanic trajectories. As such, these provided a useful constraint upon the baseline for the modelling studies. In contrast,'* (Originally P9 L 23-25), and add *'Despite this, four of the six…'* on P11 L12.

Reviewer: P9 L 7. Kim 2010 is nearly a decade old data. This is not recent and this should not be used as a justification at all. Things have changed a lot in China in the last decade.
Response: We agree that Kim *et al.,* 2010 is unlikely to reflect more recent Chinese emission trends. In addition, we also find several other studies reporting a stronger correlation between HFC-125 and HFC-32, which suggests that China does emit (and hence consume) significant quantities of R-410A. Hence, we remove *'In a recent study, Kim et al., 2010 reported a similarly weak relationship for measurements representative of Chinese emissions, suggesting that the two largest Asian economies are yet to adopt the commonly used refrigerant blend R-410A.'* (Originally P9 L7). In its place, we add the lines: *'Conversely, atmospheric measurements from China are consistent with widespread use of R-410A after 2010 (Li et al., 2011, Yao et al., 2012, Wu et al., 2018), perhaps suggesting that India lags behind China in the uptake of the HFC blends designed to replace HCFC-22.'* (P10 L20).

Reviewer: L 24 I cannot follow the argument here. Why should F-134a increase in the canister? If, then it would decrease and why only F-134a should be affected? I would simply delete this whole argument.

Response: We remove *'Several possible explanations exist for these elevated measurements: 1) Flasks collected over the Arabian Sea were compromised due to long storage times (over 1 month) at temperatures exceeding 40 °C before transport back to the UK for analysis. Long-term tests on the stability of HFC-134a at these temperatures have not been conducted; 2) the enhancements were the result of ship-borne emissions from the Indian Ocean. These flights were at low-altitude (0.01 – 0.8 km) and could have resulted in the measurement of sporadic emissions from ship-based air conditioning systems.'*

In its place we discuss only the possibility that the enhancements are the result of sporadic shipping emissions - *'One possible explanation for enhancements only being observed in HFC-134a over the Arabian Sea is that they are the result of sporadic emissions from ship-based air-conditioning systems, since all Arabian Sea samples were collected at low altitude (0.01 - 0.8 km).'* (P11 L13)

Reviewer: P12 L 1ff. The section about HFC-23 should be under the heading of HFC-23 below.
Response: We move the paragraph discussing the HFC-23 emissions total (Originally P12 L1-3) to the beginning of subsection 3.3, *'India's HFC-23 emissions and the Clean Development Mechanism'*.

Reviewer: L 26 growth? It can be also a decrease, maybe it is development in India's. . .
Response: P13 L2 has been re-structured to clarify our point that with a mandate to use an abatement system, India's future HFC-23 emissions may not mirror possible changes in the total volume of HCFC-22 produced. It now reads *'With such systems in place, possible future growth in India's HCFC-22 production rate might not result in increased emissions of HFC-23.'* (P14 L17)

Reviewer: P20 Figure 2: looking at the high baseline for HFC-32. Is this reason why the HFC-32 emissions are so low? If so that should definitely be corrected.
Response: We believe the reviewer is referring to Figure 5, which shows the derived baseline for each gas in the inversion. The baseline is not high for HFC-32 throughout the period. There is one section between flights 7-8 where the derived baseline is slightly higher than the mean but is still consistent with the uncertainty on the measurements. The majority of the time, the derived baseline in the model matches very well with the baseline measurements. Figure 2 shows HFC-32 measurements alongside data from Mace Head and Cape Grim. The baseline values are between the two sites, as expected (and similar to many of the other gases).

Reviewer: P26. Use the SPARC update for lifetimes.
Response: All atmospheric lifetimes are now taken from the 2018 Scientific Assessment on Ozone Depletion (Engel et al., 2019).

Reviewer: P28 Potential mistakes in the table. I hope I saw all but please check. This should not be like that at all! Potentially wrong: CFC-11 target (T) is it really 103? Not 101? Qualifier (Q) is it really 105, not 103 Potentially wrong: CFC-113 T: 151? Q 153? 141b Q wrong HFC-32 T wrong.
Response: We thank the reviewer for pointing out the following errors: HFC-32 target was incorrectly quoted as 23 and has been changed to 33; HCFC-141b was incorrectly quoted as 61 and has been changed to 101. The target and qualifier ions were switched for HFC-23 in order to reflect the correct ion hierarchy. Other *m/z* values are correct and with the exception of HCFC-141b, follow the work by Miller *et al.,* 2008. Upon consideration of similar publications and to simplify the table, we have removed the specific ions, leaving just the *m/z* values for each gas.

Responses to Review 2, acp-2018-1146

Reviewer: My main concern about the work is the extrapolation to annual emissions of data spanning only several weeks in one limited region of India. The author's assert that the emissions should be reasonably stable over a long period of time, but provide really no evidence that this is true. If emissions are largely from manufacturing, there can be significant variations in emissions from production facilities. Also, as the authors note, some unexpected seasonality has been observed. While error analysis is a significant part of the modelling procedure, there appears to be no estimate of additional uncertainty related to extrapolation of the short and regionally limited data set to annual and national emissions. I would like to see some clearer statement about the overall uncertainty that the authors can ascribe to the national emissions from this extrapolation. Or provide some clear caveat that, "if the emissions calculated for this time period could be scaled uniformly, then the annual emissions would be . . .. . ."

Response:

We discuss below (1) the role of production, (2) the extrapolation of emissions estimates from June-July 2016 to an annual average and (3) the extrapolation of emissions from Northern-Central India (NCI) to a national total:

1) In 2016, the only ozone-depleting refrigerant India produced was HCFC-22 (UNDP, 2013). Information from individual manufacturers suggests that HFC-134a and HFC-32 were produced by a single company in 2016 (http://www.srf.com/pdf/media/press/SRF%20Press%20Release_Refigrants02November.pdf). With the exception of HFC-23, whose predominant source is the production of HCFC-22, and HFC-32/CTC and potentially MCF, whose emissions we find are likely to mainly be from production, emissions from production are expected to be significantly smaller than emissions due to consumption for the other gases (Wan et al., 2009, McCulloch et al., 2003). We also discuss in the text on P13 L2 that there could be sporadic sources of HFC-125 in addition to widespread consumption. We therefore make the following changes to the text:

    a. Reports submitted by India's HCFC-22 manufacturers under the Clean Development Mechanism (CDM) suggest that in previous years, production of HCFC-22 (and therefore emissions of HFC-23, assuming immediate venting) did not vary significantly by month. However, there is no such evidence for 2016, and fluctuations in production rate could cause variability in annual HFC-23 emissions not captured in our estimate for June – July. We therefore add the following to P13 L24: *'Emissions of HFC-23 are linked to production of HCFC-22 and could vary in time due to unforeseen facility downtime or fluctuations in demand for HCFC-22. Based on data reported under the CDM (https://cdm.unfccc.int/Projects/registered.html), there is some evidence to suggest that bi-monthly HCFC-22 production rates have, in previous years, remained relatively constant over the course of any given year. However, these reports do not extend to 2016. While the proceeding discussion assumes that our estimate is representative of an annual total, further measurements are required to fully evaluate any short-term variability in emissions of HFC-23.'*

    b. We were unable to find any information regarding the production rate of HFC-32 and therefore add the following caveat regarding HFC-32. To P13 L20 we add: *'In addition, given emissions from production could vary in time (e.g. due to facility down-time), our emissions estimate for this gas should be considered representative of the measurement period.'*

c. We add the following (in bold), P7 L16: '*Due to sampling by aircraft, our estimates are likely to be representative on a regional-scale for gases that have sources that are widespread and do not vary significantly in time throughout the measurement period. These characteristics are thought to be true for most gases studied here. With the exception of HFC-23, HFC-32, CTC, MCF and chloroform, emissions* ***of the other gases*** *are expected to be dominated by sources linked to consumption* ***(Wan et al., 2009, McCulloch et al., 2003)****, as opposed to production.* ***Production could have short-term variations in emissions rate due to, for example, facility down-time. We also discuss below that some caution must be made in the interpretation of HFC-125 emissions.***'

2) Seasonal variations in emissions rate have been reported for two of the gases discussed in our manuscript, HCFC-22 and HFC-134a. However, in the absence of long-term datasets from the Indian subcontinent, quantification of the magnitude of seasonality is not possible. Xiang et al., 2014 estimated that global emissions of HCFC-22 and HFC-134a are two and three times, respectively, larger in summer than winter due to changes in ambient temperature and air conditioner usage. India's average temperatures do vary by season, with a minimum in winter (January-February, 22.25°C (Indian government statistics)) and a maximum in early Spring (March-May, 28.86°C). In comparison to some of the regions discussed in Xiang et al., (USA, Western Europe), the seasonality is reduced, however we agree with the reviewer the need to discuss this further. We add the following statements:

a. To P7 L3 we add: '*While the estimates presented here represent emissions over a two-month period, they are likely to be consistent with annual emissions for gases that are not expected to have significant seasonality in India.* ***Seasonal variations in emissions have been observed in HCFC-22 and HFC-134a in Western Europe and North America (Xiang et al., 2014), with summertime emissions that are two and three times larger than wintertime emissions for the two gases. The authors attribute this seasonality to increased vapour pressure in sealed refrigeration/air-conditioning systems as a result of higher ambient temperatures, and to increased use of such systems during summer months. While India's emissions of these gases could exhibit some seasonality, is not possible to estimate the magnitude of this seasonality without long-term observations from the Indian sub-continent. Our estimates for HCFC-22 and HFC-134a should be considered representative of June-July 2016 until long-term studies are conducted. Biogenic sources of chloroform have also been shown to exhibit seasonality (Laturnus et al., 2012), yet emissions from anthropogenic activities (e.g. use as a feedstock) are not likely to vary by season. No such seasonality has been reported for any of the other gases discussed here.***'

b. To P12 L9 regarding HCFC-22, we add '*Estimating seasonal variations in emission rate for India is not possible without long-term observations. Hence, our estimate for this gas should be considered representative of the measurement period.*'

c. To P12 L24 regarding HFC-134a, we add '*Previous studies reported seasonality in emissions of HFC-134a from Western Europe and North America. Without long-term measurements to quantify this seasonality in India, our emissions rate should be considered representative of the measurement period.*'

3) Quantifying the uncertainty due to scaling emissions from Northern-Central India to a national total by population requires additional measurements from southern India. Several previous studies (e.g. Barletta et al., 2011, Li et al., 2005, Stohl et al., 2009) have used similar methods to scale smaller regions into national totals using population for the gases studied here. In each of these studies, the population of the study area was considerably smaller than the national population. In our study, our NCI domain accounts for approximately 72% of India's total population. Regardless of previous publications, however, we agree with the reviewer that there will be uncertainty when performing any extrapolation.

   We now add to P6 L26: '*Emissions were aggregated into totals for the northern-central India (NCI) region (Fig. 1), which contains 72% of India's population, and then extrapolated to a national total for all gases besides HFC-32***, CTC, MCF and chloroform. The sources of the other gases except HFC-23 are refrigeration, foams, aerosols and landfills, for which we assume population to be a reasonable proxy for scaling emissions, however we are not able to quantify the uncertainty associated with extrapolating to a national total without additional measurements.***'

Reviewer: Along the same lines, it is unclear to me how uncertainties in the boundary conditions contribute to the final estimate and its uncertainty, and what might be the effect of emission plumes from beyond the Indian borders on the overall estimate of Indian emissions. My understanding is that the boundaries represent some broad regional average from a 12-box model. Would concentrated emissions from Pakistan or East Asia influence the estimates of emissions from India?

Response: The boundary conditions are estimated in the inversion on a domain that is larger than the region for which emissions are presented. We now add to the Supplement (Fig S1), the average sensitivity map over the full NAME domain. We described the domain on P5 L15, but now add reference to Fig S1: 'The model domain spanned from 55 – 109°E and 6 – 48°N up to 19 kilometres altitude (Fig S1).'

Our inversions are therefore run on a much larger domain than what is shown in Figure 1 (which is curtailed for India for presentation) and therefore includes the effect of emissions from countries outside of India. However, in general these outer regions are sufficiently far from the measurements, that their emissions do not contribute significantly to the mole fraction enhancements over background in Northern-Central India. The NAME sensitivity maps show the significant drop off in sensitivity in these other countries. For example, there is very little sensitivity to East Asia/Pakistan emissions in India at this time of year.

The 12-box model only provides a priori values for the boundary conditions on each horizontal boundary of the full NAME domain. Adjustments to these boundary conditions are then solved for in the inversion to match the 'baseline' mole fractions in the measurements. Any uncertainties in the estimation of the boundary conditions would be absorbed into emissions estimates of the outer regions of the inversion domain by design.

Reviewer: Further, it was unclear how (if) the Mace Head and Cape Grim measurements were used in the model analysis, or were just used to represent "typical" NH and SH halocarbon levels.
Response: Our paper uses the Mace Head and Cape Grim measurements in Figure 2 to represent typical northern and southern hemisphere baseline mole fractions, providing a useful comparison to our India flask data. These datasets are mainly used visually and are not used directly in the inversion. However, they are used indirectly in that measurements from these sites were used to

derive the modelled semi-hemispheric mole fractions with the AGAGE 12-box model, which were ultimately used to estimate a priori boundary conditions for the inversion (see previous comment).

Reviewer: P 3, L 33. Since there may have been some contamination in a few samples, I wonder how long the samples were stored after cleaning and before use on the flights. The note about storage in rooms without air conditioning is relevant for these measurements, but evacuated or even pressurized samples in a container that could get very toasty might also lead to artefacts in canisters with small leaks.
Response: Evacuated flasks were stored for up to 2 months prior to filling in the University of Bristol lab, where there is no air conditioning. In addition, the Medusa GCMS instrument measures a wide range of halocarbons and hydrocarbons, and of the anthropogenic species, significant enhancements in the Arabian Sea samples were only observed for HFC-134a. The expectation is that a leak in one or more of the sample flasks would result in enhancements of multiple species. To the best of our knowledge, there have been no comprehensive studies related to the stability of HFC-134a at high temperatures (in stainless steel flasks). However, there are other AGAGE stations that operate in tropical climates. Stainless steel calibration cylinders are sent via non-air-conditioned shipping containers to the AGAGE Barbados site. While the site itself is air-conditioned, this unit has failed on multiple occasions, resulting in lab temperatures that are similar to those in India. Despite this, no issues have been reported for HFC-134a. We therefore think it is more likely that those samples (which all occurred over the Arabian Sea) are picking up ship-based emissions.

Reviewer: P4, L 16. Just because I am curious about statistical calculation, could you describe how you calculated and report the overall standard deviation from triplicate sample measurements? When measurement precisions are shown are these 1 or 2 std deviations?
Response: The measurement precisions are one standard deviation of the triplicate flask analyses. The sample volume (1.75 L) was reduced in comparison to the analytical set-up described in Miller et al. 2008 (2 L), to allow for triplicate analyses to be conducted.

Reviewer: P6, L10-15. These few lines contain some assumptions that could contribute in some unknown way to the error of the method. As noted, I'd like to have some quantitative estimate of the error. E.g., "climate may minimize this", or "estimates are likely to be representative" or "characteristics are thought to be true".
Response: See response to comment 1.

Reviewer: P8, L 16 – 18. Here is where I am not sure about the use of Cape Grim to represent the conditions of the southern model boundary, or the 12-box average. I wonder if the southern boundary (from either source) might overestimate the cleanliness of the regional "unperturbed" Indian background.
Response: The Cape Grim data referred to here (originally P8 L16-18) is used visually to represent a southern hemispheric baseline for reference in Figure 2. Cape Grim data is indirectly used in the 12-box model inversion to estimate a 0-30°S semi-hemisphere value, which is then used as the a priori boundary condition for the southern inversion domain boundary. It is important to note that these a priori values are then adjusted in the inversion because offsets to each boundary are additional parameters in the inversion. They are adjusted to match the mole fractions of the "baseline" data in the measurements. We describe on P9 L4 that '*In addition to emissions parameters, a decomposition of the a priori boundary conditions, represented as offsets to the curtains in the four directions, were also solved for in the inversion.*'

Reviewer: P9, L1. And Figure 3. While there is some general correlation observed, a correlation coefficient of 0.53 is a weak argument to support common sources. There is significant variability that suggests a variety of different sources (for these and other gases), and significant variability

from possibly sporadic point sources. It is this level of variability that causes me concern about extrapolation to the whole year.
Response: An R-value of 0.53 suggests that HCFC-22 and HFC-134a share at least some common sources, or source regions, however, we acknowledge that there could be differences as well. Our work suggests that India is in a transition period, whereby HCFCs are being replaced by HFCs. In 2016, there were still significant emissions of both. However, the rate of uptake of this transition is likely to vary by region and usage, and therefore may not be uniform across all of Northern-Central India. This would contribute to a lower correlation coefficient.

We now add to P10 L11, '*It is likely that these gases share a range of common sources, including use in India's largest refrigeration and air-conditioning sector, stationary air-conditioning (Purohit et al., 2016),* ***though the rate of transition from HCFC to HFC could vary by region.***' We also reword P10 L13 so that is now reads '*We find a significant (R = 0.53, Fig. 3) relationship between HFC-134a and HCFC-22 mole fractions, consistent with* ***some*** *co-located sources.'*

While there are instances whereby an enhancement in HCFC-22 is not matched by an enhancement in HFC-134a (or vice versa), this does not necessarily mean that the sources of these gases are sporadic in time. For both gases, the model fit is good (Fig 5). Since the model assumes that emissions are constant over the measurement period, a good fit likely means that the emission model (constant emission rate) is able to simulate observations well. An example where this is not the case is HFC-125, and we discuss the fact that there could be due to sporadic sources on P13 L2.

Reviewer: P9, L 29. I think the author's aren't really talking about stability of HFC-134a, but potential for leakage and artefacts, either before or after sampling (most likely before).
Response: See response to comment 4 for discussion of potential artefacts or instability of HFC-134a at high temperatures.

Reviewer: P13, L7. I don't think that % of global emissions are expected to scale with just population, so India's 17.7% of world population wouldn't necessarily imply anything about halocarbon emissions.
Response: We had initially included this as a reference point, but we agree that one wouldn't necessarily expect global emissions to distribute according to population because of differences in Article 5 and non-Article 5 nations, production pathways, etc. We now remove any reference to the 17.7% of the global population in the Results and Discussion. However, we continue to use it to create a priori CFC emissions (scaling the global emissions from the 12-box model) as it is the best guess we have. We amend the text on P7 L26, *'To estimate a priori total emissions over India, we scaled an estimate of 2016 global emissions derived using the AGAGE 12-box model (an extension of Rigby et al. (2014)) by population, though CFC emissions are not necessarily expected to distribute globally according to population due to differences in Article 5 versus non-Article 5 country emission trends, amongst other factors.'*

Reviewer: Data availability. I would like to be able to examine the data used in this paper, but I didn't see the data availability and source listed.
Response: The data has now been uploaded to CEDA, and a link is provided in the data availability section.

Reviewer: Title: I agree with the suggestion of the first reviewer to include ".....from airborne measurements" in the title.
Response: The title of the manuscript now reads *'Emissions of halocarbons from India inferred through atmospheric measurements'*.

Responses to Review 1, acp-2018-1287

Reviewer: My main comment in the quick report was: What sets this manuscript apart from its companion paper (acp-2018-1146, Emissions of CFCs, HCFCs and HFCs from India). Both report synthetic halocarbon measurements from the same campaign which are even shown to partly correlate with each other due to similar sources.
Response: As per the author notes above, the two manuscripts have now been merged and are presented as a single study.

Responses to Review 2, acp-2018-1287

Reviewer: P 1 Line 8: This has only been 1 month of measurements not 2.
Response: This line was removed upon merging.

Reviewer: P 2 Line 8: There have been updates to this numbers in Carpenter et al. (2014) and Liang et al. 2018.
Response: This line was removed upon merging. All lifetimes, ODP and GWP values are now taken from the 2018 Scientific Assessment on Ozone Depletion (Engel et al., 2019).

Reviewer: P 2 Line 17: ODPs
Response: This line was removed upon merging.

Reviewer: P 2 Line 19: What about the new Chapter 1 of the Ozone Assessment (Engel and Rigby, 2019).
Response: All lifetimes, ODP and GWP values are now taken from the 2018 Scientific Assessment on Ozone Depletion.

Reviewer: P 2 Line 21 and 22: Hossaini et al and Fang et al is plural therefore, show and estimate.
Response: This line was removed upon merging.

Reviewer: P5 L25ff. Somehow it is unusual to use different a priori estimates for the individual compounds. Especially questionable in this respect is the use of top-down estimates as an a priori which should be independent of top-down estimates. I suggest that you use the AGAGE-12-box based method for all compounds.
Response: Different methods for compiling prior estimates for each gas were used in order to incorporate the most relevant information for each gas. However, given the large uncertainty assigned to prior in each instance, the absolute magnitude has very little influence on the posterior solution. It is also worth noting that the AGAGE 12-box model is itself a top-down estimate but is based upon independent atmospheric measurements. The main concern raised, which is of lack of independence in the prior, is not an issue here. No prior estimates based on top-down numbers have used any measurements from this study.

Reviewer: P9 L13 The focus on chloro-alkali plants is a misinterpretation of the literature. It is the total of the production of chlorine related products (chloro-alkane production and chloro-alkali plants). Citation from the conclusion of Hu et al.; Our findings suggest that the majority of US CCl4 emissions could be related to industrial sources associated with chlorine production and processing.
Response: Hu et al., conducted a Bayesian Information Criterion (BIC) analysis to determine the most likely sources of CCl4 in the US. They 'suggest that the distribution of derived posterior emissions is more consistent with that of industrial sources reported by the US EPA TRI (particularly chloralkali production plants)', though we accept that they do not exclude chloromethane production and that this industry is a likely contributor. We therefore modify P15 L16 to include other industrial sources – it now reads '*Ongoing US emissions were attributed to industrial sources, particularly chlor-alkali plants, which differs from our finding that CTC emissions in India do not correspond with known locations of chlor-alkali production.*'

Reviewer: P9 L16ff What about the correlation of CCl4 with CHCl3. If there is co-production with CH2Cl2, there should also be co-production with CHCl3, please discuss.
Response: The correlation coefficients for DCM vs. CTC and chloroform vs. CTC were both small (i.e., less than 0.2). Chloromethane manufacture is a source of all three of these gases, however, individual plants are likely to produce each component (DCM, chloroform and CTC) at a ratio unique

to that facility. In addition, we now show (P15 L6) that India likely produces more CTC than it emits. Some of this is likely to be consumed by the divinyl acid chloride (DVAC) industry. While we do not know the locations of factories producing DVAC, it is unlikely that all are co-located with chloromethane facilities. Similarly, DCM has a wide range of sources, from solvent use to foam blowing, which are not expected to correlate with the known sources (chloromethane manufacture, DVAC industry) of CTC.

Reviewer: P11. L13 . . .long-lived chlorocarbons. . .
Response: This line was removed upon merging.

Reviewer: P22. Table 2. The new Ozone Assessment has the lifetime of CCl4 as 32 years. Please correct and cite accordingly.
Response: All lifetimes, ODP and GWP values are now taken from the 2018 Scientific Assessment on Ozone Depletion, hence the lifetime of CCl4 has been updated to 32 years accordingly.

Emissions of halocarbons from India inferred through atmospheric measurements

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Abstract. As the second most populous country and third fastest growing economy, India has emerged as a global economic power. As such, its emissions of greenhouse and ozone-depleting gases are of global significance. However, unlike neighbouring China, the Indian sub-continent is very poorly monitored by existing atmospheric measurement networks. India's halocarbon emissions (here defined as chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs) and chlorocarbons) are not well-known. Previous measurements from the region have been obtained at observatories many hundreds of kilometres from source regions, or at high altitudes, limiting their value for the estimation of regional emission rates. Given the projected rapid growth in demand for refrigerants and solvents in India, emission estimates of these halocarbons are urgently needed to provide a benchmark against which future changes can be evaluated. In this study, we report the first atmospheric-measurement derived halocarbon emissions from India. Air samples were collected at low-altitude during an aircraft campaign in June and July 2016 and emissions were derived from measurements of these samples using an inverse modelling framework. These results were evaluated to assess India's progress in phasing out ozone-depleting substances under the Montreal Protocol. Our combined CFC estimates show that India contributed 54 (27 – 86) Tg CO_2eq yr^{-1}, and HCFC-22 emissions at 7.8 (6.0 – 9.9) Gg yr^{-1} were of similar magnitude to emissions of HFC-134a (8.2 (6.1 – 10.7) Gg yr^{-1}). We estimate India's HFC-23 emissions to be 1.2 (0.9 – 1.5) Gg yr^{-1} and our results are consistent with resumed venting of HFC-23 by HCFC-22 manufacturers following the discontinuation of funding for abatement under the Clean Development Mechanism. We report small emissions of HFC-32 and HFC-143a and provide evidence to suggest that HFC-32 emissions were primarily due to fugitive emissions during manufacturing processes. Lack of significant correlation among HFC species and the small emissions derived for HFC-32 and HFC-143a indicate that in 2016, India's use of refrigerant blends R-410A, R-404A and R-507A was limited, despite extensive consumption elsewhere in the world. We also estimate emissions of the regulated chlorocarbons carbon tetrachloride and methyl chloroform from Northern and Central India to be 2.3 (1.5 - 3.4) Gg yr^{-1} and 0.07 (0.04 - 0.10) Gg yr^{-1} respectively. While the Montreal Protocol has been successful in reducing emissions of many ozone-depleting substances, growth in the global emission rates of the unregulated very short-lived substances poses an ongoing threat to the recovery of the ozone layer. Emissions of dichloromethane are found to be 96.5 (77.8 - 115.6)Gg yr^{-1} and our estimate suggests a 5-fold increase in emissions since the last estimate derived from atmospheric data in 2008. We estimate perchloroethene emissions from India and chloroform emissions from Northern-Central India to be 2.9 (2.5 - 3.3) Gg yr^{-1} and 32.2 (28.3 – 37.1) Gg yr^{-1} respectively.

1 Introduction

Chlorofluorocarbons (CFCs), carbon tetrachloride (CTC) and methyl chloroform (MCF) were used widely around the world for refrigeration, air-conditioning, foam blowing and solvent applications (Montzka et al., 1999), until they were found to deplete stratospheric ozone (Molina and Rowland, 1974; Engel et al., 2019). These species were thus regulated under the Montreal Protocol on Substances that Deplete the Ozone Layer. The adoption of the Montreal Protocol and its amendments subsequently led to a marked reduction in emissions of ozone-depleting substances (ODSs) (Engel et al., 2019). However, emissions are expected to continue, particularly from developing (Article 5) nations (Vollmer et al., 2009; Wan et al., 2009), predominantly from banked sources such as refrigerators and rigid foams (Vollmer et al., 2009), and as fugitive emissions from industry (Sherry et al., 2018).

While the emissions of many ODSs are declining, emissions of some are at odds with expectations. Emissions of CFC-11 have been shown to be increasing since 2013 (Montzka et al., 2018) and evidence suggests new production from China which has not been reported to the United Nations Environment Programme (UNEP) (Rigby et al. (2019), in press). While ‘bottom-up’ emissions of CTC, estimated from reported consumption for feedstock use, are small (1 – 4 Gg yr^{-1} (Montzka et al., 2011)), ‘top-down’ studies, based on atmospheric observations, suggest actual global emissions still exceed 30 Gg yr^{-1} (Chipperfield et al., 2016; Liang et al., 2014; Lunt et al., 2018).

As a consequence of the Montreal Protocol, emissions of the first-generation of CFC replacements, the hydrochlorofluorocarbons (HCFCs), species with similar thermodynamic properties to CFCs but reduced ozone-depletion potentials (ODPs), increased considerably in the 1990s and 2000s (Montzka et al., 2009). Because HCFCs still have non-zero ODPs, they were subsequently also regulated under the Copenhagen Amendment to the Montreal Protocol in 2004. Article 5 countries are still permitted to emit HCFCs but began their HCFC phase-out in 2013, with reduction targets outlined by the HCFC phase-out management plan (HPMP (UNDP, 2013)). Recently, it was reported that global emissions of the three major HCFCs (HCFC-22, HCFC-141b and HCFC-142b) had stabilised or were decreasing, largely due to decreasing emissions from the developed world (Montzka et al., 2014; Simmonds et al., 2017).

Following regulation of HCFCs, the second-generation replacements, hydrofluorocarbons (HFCs) were adopted because they do not appreciably deplete stratospheric ozone. However, their high Global Warming Potentials (GWP, Table 1) mean that HFCs contribute to global climate change, leading to recent efforts to reduce consumption. The 2016 Kigali Amendment to the

Montreal Protocol set out targets for a gradual phase-down of HFC production and consumption. The first cuts by most Article 5 countries will not be required until 2024 and a small number of these countries will not be required to freeze emissions until 2028. Except for HFC-23 (Simmonds et al., 2018), whose emissions are a by-product of HCFC-22 production, and HFC-152a, whose emissions have stabilised since 2010 (Simmonds et al., 2016), global emissions of all major HFCs were rising until at least the end of 2016 (Simmonds et al., 2017).

While CTC and MCF are now regulated under the Montreal Protocol, very short-lived substances, such as the chlorocarbons dichloromethane (DCM), perchloroethene (PCE) and chloroform, were not considered a threat to stratospheric due to their short atmospheric lifetimes (Table 1) and thus are not regulated. However, recent studies have shown that the rapid recent growth in global emissions of DCM and chloroform has the potential to delay the recovery of the Antarctic ozone-hole (Hossaini et al., 2017; Fang et al., 2018). Hossaini et al. (2017) estimated global DCM emissions to be ~0.6 Tg yr^{-1} in 2004, which rose to over 1.1 Tg yr^{-1} by 2014. If this trend continues, Hossaini et al. (2017) shows that DCM emissions alone could lead to a delay in the recovery of the Antarctic ozone-hole by 17 - 30 years. Likewise, Fang et al. (2018) estimates that continued growth in global emissions of chloroform could result in a further delay in ozone layer recovery of 6 – 11 years.

In certain regions of the world, large-scale convective systems provide an efficient route for the transport of short-lived chlorocarbons to the stratosphere without being substantially removed in the troposphere. South Asia's monsoon systems, similar to those over eastern Asia, provide one such pathway (Fadnavis et al., 2013; Randel et al., 2010). Brioude et al. (2010) show that short-lived chlorocarbons emitted from South Asia have ODPs up to 8 times greater than those emitted from elsewhere in Asia, and 22 times greater than emissions from Europe.

India, an Article 5 country under the Montreal Protocol, ratified the Protocol in 1992. A complete phase-out of CFCs, CTC and MCF was mandated in India by 2010. Except for use in metered dose inhalers, which ceased in 2010, India reported a complete phase-out of both the production and consumption of CFCs in 2008 (UNDP, 2013). Emissions from existing banks, such as old refrigeration appliances, are however, likely to persist. Following phase-out of CFCs, India was required to reduce emissions of HCFCs. Under Stage I of the HPMP, production and consumption of HCFCs for dispersive use was designated to be frozen by January 1st, 2016, followed by complete phase-out by 2040. At the 19th Meeting of the Parties in 2007, India agreed to an acceleration of this schedule. Under Stage II of the HPMP, India agreed to freeze its consumption of HCFCs at the base level (2009/10 average) by 2013, followed by a 10% reduction (relative to the base level) by 2015 and a complete phase-out by 2030. In 2016, India adopted the Kigali Amendment, under which it will also begin to phase-down its production and consumption of HFCs. However, its developing status means it will not be required to make its first reductions until 2028, and in the meantime, India's demand for HFCs is expected to rise dramatically (Purohit et al., 2016).

With a population exceeding one billion and a rapidly expanding economy, India's halocarbon emissions are expected to have global significance. Based on inferred consumption trends, Velders et al. (2015) estimated that India will emit 400 Tg CO_2eq yr^{-1} of HFCs in 2050, a 67-fold increase over 2016 emissions. However, little else is known about India's emissions. Estimates from bottom-up, inventory-based methods have only been made for a subset of HFCs (HFC-134a, HFC-152a and HFC-23) in India and only up to 2010 (Garg et al., 2006; Ministry of Environment, Forest and Climate Change, 2012, 2015). With the exception of DCM, for which Leedham Elvidge et al. (2015) estimated India's emissions to be 20.3 (15.8 – 24.8) Gg

yr^{-1} in 2008, emissions of these gases have never been estimated for India through regional ‘top-down’ or inverse modelling approaches that use atmospheric mole fraction measurements to infer surface fluxes. However, top-down methods have been applied elsewhere in Asia (Palmer et al., 2003; Yokouchi et al., 2005; Kim et al., 2010; Saikawa et al., 2012; Stohl et al., 2010; Lunt et al., 2018).

Previous studies in other countries have shown that there can be large discrepancies between national inventories of halocarbons and those inferred from atmospheric observations (Graziosi et al., 2017; Lunt et al., 2015; Say et al., 2016). Therefore, this dual quantification approach has been highlighted by many organizations as being beneficial for accurate and transparent greenhouse gas reporting (Leip et al., 2018). In this study, we present the first top-down estimates of India’s halocarbon emissions and provide a 2016 benchmark, which is critical for evaluating future policy changes surrounding India’s halocarbon emissions.

2 Materials and Methods

2.1 Collection and analysis of air samples

Atmospheric samples were collected in evacuated 3 L stainless steel electro-polished flasks (SilcoCan, Restek, USA) aboard the UK’s FAAM (Facility for Airborne Atmospheric Measurements) BAe-146 research aircraft. In total 176 samples were collected over 11 flights conducted between the 12th June and 9th July 2016 (Table 2). On nine of these flights, samples were collected over northern India at altitudes ranging predominantly between 0 – 1.5 km (Fig. 1). Air was drawn through a forward-facing air sampling pipe on the exterior of the aircraft and pressurised into the sample flasks using a metal bellows pump (Senior Aerospace PWSC 28823-7). Sample flasks were evacuated to 1e^{-5} psig prior to each flight. Before sample collection, the lines within each sample case were flushed with ambient air for a minimum of one minute. Sample flasks were filled to a maximum pressure of 41 psig, giving a usable sample volume of 9 L at atmospheric pressure. Sample filling typically varied between 25 - 60 seconds in duration, depending on altitude (equivalent to ~7 km of flight track at average cruise velocity). Flasks were filled at regular intervals during each flight (interval dependent on flight length). When not in use, flask samples were stored in a container with no air-conditioning, to eliminate the risk of sample contamination from leaking air-conditioning refrigerant. None of the gases discussed here were present on the research aircraft itself, and the laboratory at the University of Bristol does not contain a HFC filled air-conditioning unit. Apart from flasks collected over the Arabian Sea, samples were transported from India to Bristol within one month of collection.

Flask samples were analysed using the Medusa GCMS analytical system, with modifications made to the analysis to account for the small volume and low pressure of the flask samples. In the set-up described previously (Miller et al., 2008; Arnold et al., 2012), atmospheric measurements were derived from 2 L samples, injected into the pre-concentration system at a flow rate of 100 $\text{cm}^3\ \text{min}^{-1}$, resulting in a total injection time of 20 minutes. For this work, each measurement was derived from three 1.75 L analyses and injected into the analytical system at a flow rate of 50 $\text{cm}^3\ \text{min}^{-1}$, resulting in a total injection time of 35 minutes. The analysis of each flask was bracketed by analyses of a quaternary reference gas, to account for short term drifts in detector sensitivity. Halocarbon mole fractions are reported relative to a set of gravimetrically prepared ‘primary’ standards (Table 3), via a hierarchy of compressed real-air standards held in 34 L electro-polished stainless-steel canisters (Essex Industries, Missouri, USA). The working (quaternary) standard was compared to a tertiary tank on a roughly monthly basis. System blanks were conducted monthly, to quantify possible interferences from system leaks and carrier gas impurities. For each gas, the ratio of target to qualifier ion(s) was continually monitored to ensure that co-eluting species did not interfere with the analyses. For each flask, measurement precision was estimated as the standard deviation of the three replicate analyses. Average measurement precisions are shown in Table 3 and are comparable to the precisions reported previously by Miller et al. (2008).

2.2 Numerical Atmospheric Modelling Environment (NAME)

A Lagrangian particle dispersion model was used to quantify the influence of surface fluxes on each atmospheric measurement. The Met Office NAME (Numerical Atmospheric dispersion Modelling Environment) model was run in backwards mode (Manning et al., 2011) to generate 30-day air histories for every minute along each flight path (each minute represents approximately 7 km of the flight track at average speed). These air histories represent the sensitivity of a measurement to fluxes from the surface (defined as 0 - 40 meters above ground level). NAME was driven using meteorological output from the operational analysis of the UK Met Office Numerical Weather Prediction model, the Unified Model, with a horizontal resolution of approximately 17 km in 2016. The model domain spanned from 55 – 109 °E and 6 – 48 °N up to 19 kilometres altitude (Fig. S1). For each flight minute, tracer particles were released at a rate of 1000 particles min^{-1} from a cuboid, whose dimensions were determined by the change in latitude, longitude and altitude of the aircraft during that one-minute period. In general, samples were collected during level sections of each flight path, minimising transport errors that could arise from releasing particles over a range of altitudes. At the boundaries of the domain, the three-dimensional location and time at which each particle left the domain were recorded to provide the sensitivity to boundary conditions.

Given the short lifetimes of DCM, PCE and chloroform there is some chemical loss during a typical 30-day simulation. Fang et al. (2018) investigated the impact of modelling short-lived substances with lifetimes of around six months over regional domains, without accounting for loss processes. Their study showed that, for sources that are within several hundred kilometres of measurement locations, as in this set-up, the decay is very small (less than 1%) over the time-scales of transport from source to receptor and can thus be neglected.

The ability of NAME to accurately simulate transport is critical for ensuring robust emission estimates. Model simulated wind direction and speed were compared to meteorological data recorded on board the FAAM aircraft (Fig. S2–S3). To ensure that transport errors had a minimal impact on the inversion, emissions derived using the complete set of atmospheric measurements were compared to those derived from a filtered dataset (Fig. S4), in which observations corresponding to periods where the NAME simulated wind speed/direction differed from the measured meteorology by more than 20% were removed.

2.3 Inverse modelling using atmospheric dispersion modelling

Our inverse method is based on the trans-dimensional approach described by Lunt et al. (2016). Emissions and uncertainties were characterized using principals of hierarchical Bayesian modelling detailed in Ganesan et al. (2014). The inverse approach solves for a parameter vector, $\boldsymbol{x}$ (including flux fields and boundary conditions), using measurement data, $\boldsymbol{y}$. In a Bayesian framework, independent prior knowledge of emissions, $\boldsymbol{x}_{ap}$, is used in conjunction with measurements to solve for a posterior emissions distribution, $\boldsymbol{x}$ using a linear model, $\mathbf{H}$ (Eq. 1).

$$\boldsymbol{y} = \mathbf{H}\boldsymbol{x} + \epsilon \tag{1}$$

$\mathbf{H}$ is a Jacobian matrix of sensitivities, here describing the relationship between changes in atmospheric mole fractions and changes in the parameter vector $\boldsymbol{x}$. ϵ is uncertainty arising from the model and the measurements. In a traditional Bayesian inversion, uncertainty in $\boldsymbol{x}_{ap}$ and the model-measurement uncertainty, ϵ, are both assigned prior to the inversion. These uncertainties are often poorly known and rely on a subjective decision by the investigator, but have been shown to significantly impact upon the derived posterior emissions (Peylin et al., 2002; Rayner et al., 1999). To minimize this impact, a hierarchical approach incorporates additional hyper-parameters, which allow for the propagation of 'uncertainties in these uncertainties' to the posterior solution.

$$\rho(\boldsymbol{x},\theta|\boldsymbol{y}) \propto \rho(\boldsymbol{y}|\boldsymbol{x},\theta) \cdot \rho(\boldsymbol{x}|\theta) \cdot \rho(\theta) \tag{2}$$

Eq. 2 is a hierarchical version of Bayes' theorem (normalizing factor not shown for brevity). In this example, the prior emissions uncertainty is governed by a hyper-parameter (θ), which has a probability density function (PDF) that is explored within the inversion. This equation can also be employed in a similar way for the model-measurement uncertainty or any other unknown parameters. The hierarchical Bayesian approach was extended to a trans-dimensional framework, in which the number and configuration of the spatial grid over which emissions were estimated were also unknown parameters, prior to the inversion. Therefore, it is largely the information content of the measurements that govern these unknown aspects. This framework has been shown to result in a more robust and justifiable quantification of uncertainties in emissions than traditional approaches.

In general, Eq. 2 is not solvable via analytical means and was estimated using reversible jump Markov Chain Monte Carlo (rj-MCMC). The rj-MCMC algorithm was used to sample 320,000 variants of the parameter space with the first 120,000 discarded as 'burn-in' to ensure that the system had no knowledge of the initial state. The remaining 200,000 samples were then used to form the posterior PDFs. In our estimates, the means of these posterior PDFs are presented, with the uncertainties represented by the 5^{th} and 95^{th} percentile values.

Emissions were aggregated into totals for the northern-central India (NCI) region (Fig. 1), which contains 72% of India's population, and then extrapolated to a national total for all gases besides HFC-32, CTC, MCF and chloroform. The sources of the other gases except HFC-23 are refrigeration, foams, aerosols and landfills, for which we assume population to be a reasonable proxy for scaling emissions, however we are not able to quantify the uncertainty associated with extrapolating to a national total without additional measurements. For HFC-23, the NCI region incorporated four of the five known manufacturing plants for HCFC-22. To estimate national emissions, we scaled the NCI total by the ratio of HCFC-22 produced at those four factories, to total production at all five (based on 2015 factory specific production statistics (UNEP, 2017)). Based on these statistics, over 98% of HCFC-22 was produced by factories residing within the NCI.

While the estimates presented here represent emissions over a two-month period, they are likely to be consistent with annual emissions for gases that are not expected to have significant seasonality in India. Seasonal variations in emissions have been observed in HCFC-22 and HFC-134a in Western Europe and North America (Xiang et al., 2014), showing that summertime emissions are two and three times larger than wintertime emissions for the two gases, respectively. The authors attribute this seasonality to increased vapour pressure in sealed refrigeration/air-conditioning systems as a result of higher ambient temperatures, and to increased use of such systems during summer months. While some degree of seasonality might be expected for India's emissions of these gases, is not possible to estimate the magnitude of seasonality without long-term observations from the Indian sub-continent. Our estimates for HCFC-22 and HFC-134a should be considered representative of June-July 2016 until long-term studies are conducted. Biogenic sources of chloroform have also been shown to exhibit seasonality (Laturnus et al., 2002), yet emissions from anthropogenic activities (e.g. use as a feedstock) are not likely to vary by season. No such seasonality has been reported for any of the other gases discussed here.

Due to sampling by aircraft, our estimates are likely to be representative on a regional-scale for gases that have sources that are widespread and do not vary significantly in time throughout the measurement period. These characteristics are thought to be true for most gases studied here. With the exception of HFC-32, HFC-23, CTC, MCF and chloroform, emissions of the other gases are expected to be dominated by sources linked to consumption (Wan et al., 2009; McCulloch et al., 2003), as opposed to production. Production could have short-term variations in emissions rate due to, for example, facility down-time. We also discuss below that some caution must be made in the interpretation of HFC-125 emissions.

2.4 A priori emissions

A priori emissions were assembled from a variety of sources owing to the limited information available for India. **CFCs**: Since a total ban on CFC production and consumption has been in place since 2010, country specific emissions/consumption data no longer exist. Despite this, studies suggest that emissions of these gases could be ongoing (Montzka et al., 2018). To estimate a priori total emissions over India, we scaled an estimate of 2016 global emissions derived using the AGAGE 12-box model (an extension of Rigby et al. (2014), see section 2.6) by population (though CFC emissions are not necessarily expected to distribute globally according to population due to differences in Article 5 versus non-Article 5 country emission trends, amongst other factors). **HCFCs**: A priori total HCFC emissions over India were based on 2015 consumption data reported by India in its HPMP Stage II Road Map report (Ministry of Environment, Forest and Climate Change, 2017). Consumption is likely an underestimate of emissions due to the presence of banked sources such as refrigerators and foams. **HFCs**: Excluding HFC-23, prior HFC emission totals for India were calculated by scaling the 2010 EDGAR v4.2 (European Commission, 2009) Asian continental total by population (with India accounting for approximately 29% of the Asian total). This was done because EDGAR does not indicate any emissions from India. For HFC-23, prior emission totals for India were based on the 2010 HFC-23 emissions reported in India's Biennial Update Report to the United Nations Framework Convention on Climate Change (Ministry of Environment, Forest and Climate Change, 2015), and extrapolated to 2016 using reported HCFC-22 production data (and assuming a constant co-production ratio) (Ministry of Environment, Forest and Climate Change, 2017). **Regulated chlorocarbons**: India's CTC emissions were estimated at 2.8 Gg yr^{-1} based on the 2014 estimate by Sherry et al. (2018). As with the CFCs, a priori MCF emissions were calculated using a population-based scaling of the global total derived using the AGAGE 12-box model, and hence estimated to be 0.3 Gg yr^{-1} in 2016. **Unregulated chlorocarbons**: For DCM, a priori emissions were from Leedham Elvidge et al. (2015), which estimated India's DCM emissions to be 20.3 Gg yr^{-1} in 2012 based on independent measurements. India's PCE emissions were from the Reactive Chlorine Emissions Inventory (McCulloch et al., 1999) and were estimated at 6.0 Gg yr^{-1}. Terrestrial chloroform emissions were taken from the AGAGE 12-box model. Using a population scaling for India and assuming that 45% of chloroform emissions (biogenic and anthropogenic) originate on land (McCulloch, 2003), India's land-based chloroform emissions were estimated at 3.0 Gg yr^{-1}. Oceanic chloroform emissions were adapted from Khalil et al. (1999), who estimated a northern hemispheric tropical ocean source of 50 Gg yr^{-1}. The ocean within our model domain was estimated to account for 8.3% of this source by area, equivalent to 4.2 Gg yr^{-1}.

No recent spatial information was available for any of the halocarbons studied here. With the exception of chloroform, for which prior emissions were distributed uniformly across both land and ocean, prior emissions totals were distributed across the model domain using the National Oceanic and Atmospheric Administration (NOAA) DMSP-OLR satellite night light data, available at 30 arc second resolution (https://ngdc.noaa.gov/eog/data/web_data/v4composites/). The night lights distribution was a useful starting point for our emissions maps, since night lights are generally correlated with population density (Raupach et al., 2010), but are also likely to include industrial sites, such as HCFC-22/chloromethane manufacturing plants. We expected the major sources of CFCs, HCFCs, HFCs and chlorocarbons to be explicitly linked to domestic and/or commercial activities, or from industries requiring a significant work-force.

For all species, the prior emissions uncertainty was described by a uniform PDF with lower and upper bounds of 50% and 500% respectively. This large uncertainty reflects the lack of detailed information available for India. Large prior uncertainties mean that our posterior emissions over the NCI are informed almost entirely by the atmospheric measurements. To confirm that our posterior estimates were independent of the spatial distribution of the prior, results derived using the night-lights data were compared to those derived from a spatially uniform prior (Fig. S4).

2.5 A priori boundary conditions

The footprints from NAME only model the emissions released within the model domain. Hence, a prior estimate of the mole fraction at the boundaries of model domain must be made and incorporated into the modelled mole fraction. Mole fraction 'curtains' of each gas were used to provide a priori information about boundary conditions (it should be noted that these boundary conditions were adjusted within the inversion). For the HFCs, mole fractions were simulated using the 3D global chemical transport model MOZART (Model for OZone and Related chemical Tracers (Emmons et al., 2010)). MOZART was driven by offline meteorological fields from MERRA (Modern Era Retrospective-Analysis for Research and Applications (Rienecker et al., 2011)). For the CFCs, HCFCs and chlorocarbons, MOZART fields were not available, and uniform curtains were assumed. The mole fraction for each curtain in each month was estimated using the AGAGE 12-box model (Rigby et al., 2014) and measurements from five baseline AGAGE observatories. For each gas, the model was used to estimate a monthly baseline mole fraction for four latitude bands. The simulated mole fraction from latitude bands 30 – 90 °N, 0 – 30 °N and 0 – 30 °S were used to assign a priori mole fractions to the northern, eastern/western and southern curtains of the model domain respectively. The boundary conditions associated with each NAME-simulated measurement were calculated by mapping the exit times and locations of particles leaving the domain to the curtains. In addition to emissions parameters, a decomposition of the a priori boundary conditions, represented as offsets to the curtains in the four directions, were also solved for in the inversion.

2.6 Global halocarbon emissions estimation

Indian halocarbon emissions were compared to global emission estimates calculated using the AGAGE 12-box model (Rigby et al., 2014), assimilating data from five remote AGAGE background sites (Mace Head, Ireland; Trinidad Head, USA; Ragged Point, Barbados; Cape Matatula, American Samoa and Cape Grim, Tasmania) following a Bayesian inversion methodology. Baseline monthly means were estimated by statistically filtering the high-frequency data (O'Doherty et al., 2001). The data were averaged into semi-hemispheres (30 °N – 90 °N, 0 °N – 30 °N, 30 °S – 0 °S, 90 °S – 30 °S) for comparison with mole fractions predicted by the AGAGE 12-box model, which resolves these four semi-hemispheres, with vertical levels separated at 500 and 200 hPa (Cunnold et al., 1983; Rigby et al., 2013). The model uses annually repeating meteorology and OH concentrations from Spivakovsky et al. (2000), tuned to match the growth rate of methyl chloroform.

Total atmospheric lifetimes (Table 1) were estimated using the halocarbon-hydroxyl temperature-dependent rate constants from Burkholder et al. (2015) (tropospheric removal) and the average photochemical model loss frequencies given in Ko et al. (2013) (stratospheric removal). A Bayesian framework was used to derive emissions from the data and the model, in which an a priori estimate of the emissions growth rate was adjusted to bring the model into agreement with the data (following Rigby et al. (2011)). The inversion propagates uncertainties in the observations through to the derived fluxes and augments the derived fluxes with uncertainties due to the lifetime and potential errors in the calibration scale. These estimates are a 2016 extension of those presented in Rigby et al. (2014).

3 Results

3.1 Atmospheric Measurements

Measurements were made from whole air flask samples collected over India during June and July 2016. Fig. 1 shows the location and altitude of these measurements along with the model-derived sensitivity of these samples to surface emissions. Mole fractions of each halocarbon measured during the campaign are shown in Fig. 2. For comparison, each gas is shown alongside baselines representative of the Northern and Southern Hemispheres. These baselines were derived from statistical fits to observations from the AGAGE sites at Mace Head, Ireland and Cape Grim, Tasmania (Prinn et al., 2018). Although all of the samples collected during our campaign were within the Northern Hemisphere, the South Asian monsoon, which occurs annually between June and September, draws air from southern latitudes, resulting in the Indian regional background being more consistent with the Southern Hemisphere at this time of year. For CFC-12 and CFC-113, owing to the decrease in emissions resulting from the Montreal Protocol, the hemispheric baselines are now similar and the difference in mole fraction between hemispheres is smaller than the average precision of our flask measurements.

Enhancements in mole fractions over the regional background form the basis for estimating regional emissions. For all species except HFC-134a, the average mole fractions of samples collected over the Arabian Sea were lower than those collected directly over NCI. Variability in the mole fraction of samples collected over NCI varied considerably by species. For CFC-11, CFC-12 and CFC-113, few pollution events were observed, and their signals were of similar size to the measurement precision. Similarly, only small enhancements were observed for HCFC-142b, suggesting its main use as a foam-blowing agent was not significant or was not widespread and thus could not be discerned in the aircraft samples.

In contrast, large enhancements in mole fraction were observed for HFC-134a and HCFC-22, suggesting that usage of these substances as a refrigerant is widespread. It is likely that these gases share a range of common sources, including use in India's largest refrigeration and air-conditioning sector, stationary air-conditioning (Purohit et al., 2016), though the rate of transition from HCFC to HFC could vary by region. We find a significant (R = 0.53, Fig. 3) relationship between HFC-134a and HCFC-22 mole fractions, consistent with some co-located sources. Large enhancements in HFC-23 mole fraction suggest that the samples were sensitive to emissions from HCFC-22 manufacturing facilities, as HFC-23 is a by-product of HCFC-22 production. The NCI region contains four out of the five Indian manufacturing facilities that were registered under the Clean Development Mechanism (CDM) (https://cdm.unfccc.int/Projects/registered.html).

Enhancements were also observed in HFC-32 and HFC-125, although the observed mole fractions for these species were not strongly correlated (R = 0.15, Fig. 3), suggesting that India is yet to adopt refrigerant blend R-410A (50% by wt. HFC-125, 50% by wt. HFC-32) on a large scale. Conversely, atmospheric measurements from China are consistent with widespread use of R-410A after 2010 (Li et al., 2011; Yao et al., 2012; Wu et al., 2018), suggesting that India lags behind China in the uptake of the HFC blends designed to replace HCFC-22, or that it has adopted lower GWP alternatives. Similar to China, HFC-125/HFC-32 measurements at Mace Head (Ireland) over the same time-period were strongly correlated (R = 0.86). All enhancements in HFC-32 are found to correspond with enhancements in DCM (Fig. 3), suggesting that India's emissions of this gas are linked to its production. The significance of this correlation is discussed further in section 3.2.3.

We found no correlation for HFC-125 and HFC-143a (R = -0.04, Fig. 3), gases whose emissions are regularly linked through the consumption of blends R-404A (52% by wt. HFC-143a, 44% by wt. HFC-125, 4% by wt. HFC-134a) and R-507A (50% by wt. HFC-125, 50% by wt. HFC-143a) (Montzka et al., 2014; O'Doherty et al., 2014). In contrast, at Mace Head, a strong HFC-125/HFC-143a correlation (R = 0.78) was observed during this time.

Evidence for widespread use of both HCFCs and HFCs and the lack of large enhancements in CFCs suggests that India's transition to first- and second-generation CFC replacements is nearing completion. However, there appears to be little evidence for the consumption of HFC blends or HFC-152a in 2016, refrigerants/propellant used extensively in the developed world (Greally et al., 2007; O'Doherty et al., 2014).

Only a small number of enhancements were observed for the regulated chlorocarbons CTC and MCF, while a large number of enhancements were observed for all three unregulated chlorocarbons. In particular, very large enhancements were found for DCM, with a maximum mole fraction of 1133 ppt (corresponding enhancement of 1120 ppt, Fig. 2). Samples collected at longitudes east of 81 °E were particularly enhanced above the baseline, suggesting that the flask samples were sensitive to regions producing/consuming large quantities of DCM as a solvent, feedstock or both.

We found a significant (R = 0.71) correlation between DCM and chloroform (Fig. 3), suggesting that these gases share some similar sources or source locations (i.e. DCM and chloroform are chloromethanes manufactured for use as feedstock gases for HFC-32 and HCFC-22, respectively). Since DCM is predominantly anthropogenic in origin, this correlation indicates that some of the enhancements observed for chloroform are from anthropogenic sources. However, we find a low correlation (R = 0.24) between HFC-23 and chloroform (Fig 3). This suggests that either fugitive losses during chloroform manufacture are not co-incident with losses during HCFC-22 production, or there are also other sources of chloroform such as biogenic sources.

During two flights (B959 and B963) conducted on the 21st and 25th of June, a small number of samples were collected over the Arabian Sea. NAME back-trajectory analysis was used to show that these samples had not interacted with any significant landmass in the 30 days prior to collection. Despite this, four of the six samples collected on these flights exhibited an elevated HFC-134a concentration, which did not correlate with any other species, including HCFC-22. One possible explanation for enhancements only being observed in HFC-134a over the Arabian Sea is that they are the result of sporadic emissions from ship-based air-conditioning systems, since all Arabian Sea samples were collected at low altitude (0.01 - 0.8 km).

3.2 Halocarbon emissions estimates for NCI and India

Mean NCI and Indian emissions estimates and the relative contributions of each gas to 2016 global emissions are shown in Fig. 4 and tabulated in Table 4 (Gg yr^{-1}) and Table 5 (Tg CO_2eq yr^{-1}). Uncertainties presented throughout correspond to the 5th and 95th percentiles of the posterior distribution.

We estimate India's 2016 CFC, HCFC and HFC (excluding HFC-32) emissions to be 54 (27 – 86) Tg CO_2eq yr^{-1}, 15 (11 - 19) Tg CO_2eq yr^{-1} and 53 (40 – 67) Tg CO_2eq yr^{-1} respectively, which correspond to 7 (4 – 12) %, 2 (1 – 3) % and 6 (5 – 8) % of global emissions. Combined emissions of regulated (CTC and MCF) and unregulated (DCM, PCE and chloroform) chlorocarbons from NCI are estimated at 11 (7 - 16) Tg CO_2eq yr^{-1} and 1 (1 - 2) Tg CO_2eq yr^{-1}, which account for 7 (4 - 10) % and 8 (6 - 9) % of global emissions, respectively. With the exception of DCM, there are no previous top-down national-scale estimates of any of these gases for India. In 2016, India's aggregated HFC emissions were approximately an order of magnitude larger than the 2016 emissions assumed by Velders et al. (2015), suggesting that future projections of India's HFC emissions could be inaccurate.

3.2.1 CFCs

Through commitments under the Montreal Protocol, India finalised its phase-out of the consumption and production of CFCs in 2010. However, residual emissions from banks (refrigerators, foams, landfills etc.) are expected to continue for several decades (Rigby et al., 2014). Our mean CFC-11, CFC-12 and CFC-113 emissions are 1.7 (0.8 – 3.1) Gg yr^{-1}, 4.1 (2.1 – 6.3) Gg yr^{-1} and 0.5 (0.2 – 0.8) Gg yr^{-1}, respectively, corresponding to 2 (1 - 4) %, 13 (7 - 20) %, and 7 (2 - 11) % of global emissions in 2016. The magnitude of the uncertainties in our CFC estimates are largely a reflection of the precision of the measurements.

Further work is needed through additional high-precision measurements, particularly for CFC-12, to narrow this uncertainty. For CFC-11, our 2016 estimate of 1.2 (0.6 – 2.2) Gg yr^{-1} suggests that NCI, the region with the majority of India's population, is unlikely to have contributed significantly to the recent rise in global emissions (an increase of 13 ± 5 Gg yr^{-1}) reported between 2013 – 2016 (Montzka et al., 2018).

3.2.2 HCFCs

There is limited information about HCFC emissions from India, with the current state of knowledge encapsulated only in reports of production and consumption (Ministry of Environment, Forest and Climate Change, 2017). We find that India's 2016 HCFC emissions are dominated by HCFC-22 at 7.8 (6.0 – 9.9) Gg yr^{-1}, and these emissions comprise only 2 (1 - 3) % of global emissions. Estimating seasonal variations in emission rate for India is not possible without long-term observations. Hence, our estimate for this gas should be considered representative of the measurement period only. Our HCFC-22 emissions are comparable in magnitude to HFC-134a and HFC-125, discussed below, suggesting that India's transition from HCFCs to their non-ozone depleting replacements is in progress. India's HCFC-22 emissions are considerably smaller than those from other nations such as China, whose emissions in 2007 were estimated at 165 (140 - 213) Gg yr^{-1} (Vollmer et al., 2009) and the USA, whose emissions in 2014 were estimated at 40.0 (34.1 – 45.8) Gg yr^{-1} (Hu et al., 2017).

Our estimates of India's HCFC-141b and HCFC-142b emissions are small (1.0 (0.7 – 1.5) Gg yr^{-1} and 0.10 (0.06 – 0.14) Gg yr^{-1}, respectively). Taken together with the small reported consumption of these gases in 2015, our results suggest that either these substances have not had widespread usage in India or that efforts have been made by India under Stage I of the HPMP to phase-out HCFC consumption in the foam sector (Ministry of Environment, Forest and Climate Change, 2017), in favour of zero-ODP alternatives (UNDP, 2013). However, without detailed emissions information from previous years, it is not possible to determine whether the latter has been in effect.

3.2.3 HFCs

India's HFC emissions are dominated by emissions of HFC-134a and HFC-125, with estimated rates of 8.2 (6.1 – 10.7) Gg yr^{-1} and 6.4 (5.2 – 7.8) Gg yr^{-1} respectively. These emissions correspond to 4 (3 - 5) % and 10 (8 - 12) % of global emissions. Previous studies reported seasonality in emissions of HFC-134a from Western Europe and North America. Without long-term measurements to quantify this seasonality in India, our emissions rate should only be considered representative of the measurement period.

There are significant discrepancies between previous bottom-up estimates and our top-down results. Garg et al. (2006) estimated Indian HFC-134a emissions to be 1.1 Gg yr^{-1} in 2005, while India reported 0 Gg yr^{-1} in 2010 in its Biennial Update Report to the United Nations Framework Convention on Climate Change (UNFCCC) (Ministry of Environment, Forest and Climate Change, 2015).While there are no top-down comparisons for 2005, our results show there could have been significant growth in emissions of HFC-134a since 2005 and/or large discrepancies between bottom-up and top-down methodologies.

Further work and additional measurements are required to better understand the non-refrigerant blend sources of HFC-125. Our results suggest a possible application of HFC-125 in India as a standalone refrigerant, or an application that is not currently for HCFC-22 replacement. Possible contributors are fire suppression, use as a solvent and the production of HFC-125 for export. While our model was able to capture most of the signals for the gases studied here (Fig. 5), it was unable to simulate some of the elevated measurements for HFC-125, indicating that in addition to widespread, constant sources, there could be point sources of HFC-125 that are episodic and difficult to resolve in a model.

We estimate India's emissions of HFC-143a to be 0.8 (0.4 – 1.2) Gg yr^{-1}, which comprise 3 (1 - 4) % of global emissions. Our low HFC-143a estimate corroborates our assertion of minimal R-404A and R-507A consumption. There are no previous estimates for Indian HFC-143a emissions.

India's HFC-152a emissions are estimated to be 1.2 (0.9 – 1.4) Gg yr^{-1}, which amount to 2 (2 - 3) % of global emissions. Garg et al. (2006) estimated India's HFC-152a emissions to be 0.04 Gg yr^{-1} in 2005 and attributed these to the glass industry. Our emission rate is comparatively large, suggesting that either there are discrepancies with inventory methodologies or that there has been substantial growth in emissions in the last decade. Regardless, these emissions are small compared to other countries, particularly China, whose emissions of HFC-152a were estimated at 16 Gg yr^{-1} in 2013 (Fang et al., 2016), and the USA, for which an emission rate of 51.5 (35.5 – 75.5) Gg yr^{-1} was estimated for 2012 (Simmonds et al., 2015).

HFC-32 emissions are estimated for NCI to be 0.44 (0.36 – 0.54) Gg yr^{-1}. All the measured enhancements in HFC-32 are correlated with enhancements in DCM, a feedstock in the manufacture of HFC-32 (Fig. 3). These measurements suggest that India's HFC-32 emissions originate predominantly from fugitive losses during the manufacturing process, rather than widespread use in a refrigerant blend. Our assertion is consistent with a previous study (Leedham Elvidge et al., 2015), which attributed growth in South Asian emissions of DCM to HFC-32 manufacture. Since our NCI HFC-32 estimate is attributed to production, we consider it to be decoupled from population density, and hence we have not scaled this value to a national total. In addition, given emissions from the manufacturing process could vary in time (e.g. as a result of facility down-time), our emissions estimate for this gas should be considered representative of the measurement period only.

3.3 India's HFC-23 emissions and the Clean Development Mechanism

HFC-23 emissions are estimated for India to be 1.2 (0.9 – 1.5) Gg yr^{-1}, which comprise 10 (7 - 12) % of global emissions. Emissions of HFC-23 are linked to production of HCFC-22 and could vary in time due to unforeseen facility downtime or fluctuations in demand for HCFC-22. However, based on data reported under the CDM (https://cdm.unfccc.int/Projects/registered.html), there is evidence to suggest that HCFC-22 production rates have in previous years remained relatively constant over any given year. While we therefore assume that our estimate is representative of an annual average, further measurements are required to fully evaluate any short-term variability in emissions of HFC-23. Fig. 6 shows that emission 'hot-spots' picked out by the inverse model are consistent with the known locations of HCFC-22 manufacturing facilities.

Between 2004 and 2013, India received substantial funding from the Clean Development Mechanism for the abatement of HFC-23 produced during the manufacture of HCFC-22. To assess the impact of the CDM on India's HFC-23 emissions, we compare our HFC-23 emission estimate to previous estimates derived from bottom-up methods (Fig. 7). Emissions between 1990 and 2005 are from Garg et al. (2006), and in 2007 and 2010 are from India's reports to the UNFCCC (Ministry of Environment, Forest and Climate Change, 2012, 2015). The reported bottom-up estimates show accelerating growth in India's

HFC-23 emissions, which increased from 0.07 Gg yr^{-1} in 1990 to 1.43 Gg yr^{-1} in 2010. It is important to note that there is a large discrepancy in emissions reported in the UNFCCC inventory and in the manufacturers' CDM submissions, suggesting inconsistencies in the two methodologies. These discrepancies highlight the value of independent top-down estimates.

Depending on the efficiency of the manufacturing process, the HFC-23/HCFC-22 production ratio can vary between 0.014 (Rotherham, 2004) for optimised processes to values in excess of 0.04 for inefficient processes (McCulloch and Lindley, 2007). The production ratio is equal to the quantity of HFC-23 produced with respect to the quantity of HCFC-22 produced and is equivalent to an HFC-23 emission ratio when no abatement technologies are implemented. Based on India's HCFC-22 production statistics and bottom-up HFC-23 emission estimates, in 2007, prior to when all five manufacturers of HCFC-22 reported the use of abatement technologies, the average production ratio was 0.031.

The Clean Development Mechanism was in operation in India between 2004 and 2013. During the period of the CDM when abatement was in use at all facilities (2009 - 2013), the average emission ratio dropped to 0 – 0.009 based on the amount of non-abated HFC-23 (i.e. emissions vented to the atmosphere) reported by the manufacturing facilities. Our top-down estimate in 2016 corresponds to an average emission ratio of 0.022. While the CDM may have been effective in reducing HFC-23 emissions, our results are consistent with resumed venting of HFC-23 by some or all manufacturers, following the discontinuation of CDM funding. In October 2016, the Indian government issued a national order requiring all manufacturers of HCFC-22 to maintain a proven abatement system and ensure capacity for the storage of HFC-23 for abatement system down-time. With such systems in place, possible growth in India's HCFC-22 production rate might not result in increased emissions of HFC-23.

3.4 Regulated chlorocarbons

We estimate CTC emissions from NCI to be 2.3 (1.5 - 3.4) Gg yr^{-1}, which accounts for 7 (4 - 10) % of global emissions in 2016. India reported that its production and consumption of CTC had ceased prior to 2016 (http://ozone.unep.org/countries/data). Since then, ongoing CTC emissions from India may not be linked to its use as solvent but may persist due to fugitive leaks during chloromethane manufacture (most notably DCM and chloroform production) and from chlorine consuming industries, such as chlor-alkali plants. India does not have any major operational facilities for the manufacture of PCE, which is another anthropogenic source of CTC (Sherry et al., 2018). As these activities are not thought to be distributed evenly with respect to population, we do not scale NCI estimates to a national total.

Sherry et al. (2018) estimated that India's chloromethane manufacturers might have produced as much as 20 Gg of CTC as by-product in 2014, with corresponding fugitive emissions of 1.8 Gg yr^{-1}, though these findings are not reflected in the UNEP reports. CTC is typically produced as a by-product of chloromethane (DCM and chloroform) manufacture at an estimated rate of 4%, and the ratio of DCM to chloroform production, while variable, typically varies from 30:70 to 70:30 (Oram et al., 2017; Sherry et al., 2018). Hence, if the total production of DCM/chloroform is known, the quantity of CTC produced may also be inferred. While we were unable to find any chloromethane production data for India, chloroform is used as a feedstock in the production of the refrigerant HCFC-22. Over 99% of chloroform produced globally is used in the manufacture of HCFC-22, with 1 kg of HCFC-22 requiring 1.5 kg of chloroform as feedstock (Oram et al., 2017). Based on an extrapolation of reported

HCFC-22 production statistics in India (available from 2006 – 2015 (UNEP, 2017)), we estimate India's HCFC-22 production in 2016 to be 55 Gg. If all chloroform produced was used for HCFC-22 manufacture, and all demand was met domestically (available data suggests India only imported ~165 tonnes of chloroform in 2016 (https://www.seair.co.in/chloroform-import-data.aspx)), we estimate that India would produce 82.5 Gg of chloroform in 2016. Based on the possible DCM/chloroform production ratios discussed above, India is estimated to have produced 117 - 275 Gg of chloromethanes, and hence 4.7 - 11.0 Gg of CTC, in 2016. Since the majority of India's chloromethane manufacture occurs within NCI, this suggests that a significant amount of CTC is either destroyed or sold for non-dispersive applications. One such application is the production of divinyl acid chloride (DVAC). Sherry et al. (2018) estimated that India's DVAC industry consumed 20 Gg of CTC in 2014.

Our posterior emissions map (Fig. 6) shows that the majority of CTC emissions originate from chloromethane manufacturing facilities, while the known locations of chlor-alkali plants do not appear to be associated with large emissions. The emissions distribution of CTC resulting from the inversion is similar to that of HFC-23. This may be because CTC is a by-product of chloromethane (i.e., chloroform and DCM) manufacture, and HFC-23 is produced during the manufacture of HCFC-22, which requires chloroform as a feedstock. The locations of the main HCFC-22 production facilities are in similar locations to the chloromethane facilities in NCI (Fig 6). India's CTC emissions remain small compared to those of eastern China, whose average emissions from 2011 - 2015 were estimated at 17 (11 – 24) Gg yr^{-1} (Lunt et al., 2018), but are of similar magnitude to those of the US, estimated at 4.0 (2.0 – 6.5) Gg yr^{-1} between 2008 – 2012 (Hu et al., 2016). Ongoing US emissions were attributed to industrial sources, particularly chlor-alkali plants, which differs from our finding that CTC emissions in India do not correspond with known locations of chlor-alkali production.

Based on its reports to the UNEP, India has not produced or consumed MCF since 2001. However, a small number of enhancements in the mole fraction of this gas suggests that sources persist. Reimann et al. (2005) proposed that factories producing HCFC-141b and HCFC-142b were possible sources of MCF in Europe, since MCF is used as a feedstock in the production of these refrigerants. However, India does not report production of either HCFC-141b or HCFC-142b (Ministry of Environment, Forest and Climate Change, 2017). Landfills are another possible source of MCF, with previous studies from other regions reporting emissions from municipal waste disposal facilities (Maione et al., 2014; Talaiekhozani et al., 2018). Therefore, the nature, location and magnitude of the sources of MCF are uncertain and we do not estimate a total for the whole of India. At 0.07 (0.04 - 0.10) Gg yr^{-1}, MCF emissions from the NCI account for 4.1 (2.4 – 5.9) % of global emissions. Despite its status as a developing country, which meant India had more time to phase-out consumption of MCF compared to developed countries, emissions from the NCI (which comprises 72% of India's population and includes several key industrial regions) are smaller than those from Europe, which were estimated to be 0.20 Gg yr^{-1} in 2012 (Maione et al., 2014). Given the continued role of MCF in estimating global hydroxyl concentrations (e.g. Rigby et al. (2017)), further long-term measurements from India are required to better understand the remaining sources of this gas.

3.5 Unregulated chlorocarbons

We estimate Indian DCM emissions to be 96.5 (77.8 - 115.6) Gg yr^{-1}, and these contribute 11 (9 - 13) % of global emissions. India's DCM emissions are small compared to the 455 ± 45.5 Gg yr^{-1} emitted from China in 2015 Oram et al. (2017). When compared to previous estimates of India's DCM emissions, our results reflect substantial growth. Leedham Elvidge et al. (2015) estimated emissions of 4.9 (2.7 - 7.2) Gg yr^{-1} in 1998, rising to 20.3 (15.8 - 24.8) Gg yr^{-1} in 2008, suggesting a 2- to 4-fold increase in emissions over that period. Our mean estimate represents an approximate 5-fold increase in emissions between 2008 and 2016. Global emissions over the same period rose from 611.5 Gg yr^{-1} to 907.3 Gg yr^{-1}, representing an increase of 295.8 Gg yr^{-1}. The growth in India's emissions over this period (48.9 Gg yr^{-1}) would therefore represent 25.8% of the global rise. The rise in India's DCM emissions could possibly be attributed to increased production of HFC-32, however, no HFC-32 production information from India is available. Our HFC-32 measurements suggest that a large proportion of the HFC-32 produced by India is exported rather than consumed.

Emissions of PCE are almost exclusively anthropogenic in origin, due to its widespread use as a chemical intermediate and general-purpose solvent. Despite classification as a hazardous air pollutant by the United States Environmental Protection Agency (EPA, 2012), PCE is used extensively in India as a dry-cleaning solvent (Srivastava, 2010). We estimate India's PCE emissions to be 2.9 (2.5 - 3.3) Gg yr^{-1}, which account for 4 (3 - 4) % of the global total. When compared to the only previous estimate of India's PCE emissions, which was calculated using bottom-up methods (3.9 Gg yr^{-1} in 1990 (McCulloch et al., 1999)), our estimate either shows a discrepancy with bottom-up inventories or a decrease in emissions since 1990. The latter would be consistent with global emissions derived using the AGAGE 12-box model (Rigby et al., 2014), which also show a decline from 124.2 (50.3 – 204.2) Gg yr^{-1} in 2006 to 82.6 (35.8 – 133.3) Gg yr^{-1} in 2016.

Because India's chloroform emissions are linked to industrial processes (chloromethane and HCFC-22 manufacture) and biogenic emissions, we do not scale to a national total. We estimate NCI chloroform emissions to be 32.2 (28.3 – 37.1) Gg yr^{-1}, and these emissions account for 10 (8 – 11) % of global emissions in 2016. However, given the large biogenic component of global emissions, the contribution of the NCI to global anthropogenic emissions may be significantly larger.

3.6 Sensitivity tests

We performed two sensitivity tests. We assessed the sensitivity of derived emissions to the a priori emissions field. We also assessed the effect of inaccurate transport modelling on derived emissions by using a second, filtered dataset, removing times where NAME wind direction and wind speed differed by more than 20% from the measured parameters. A comparison of the three posterior estimates is given in Fig. S4. For all 17 halocarbons, the three estimates are statistically consistent, indicating that our estimates were robust to the prior spatial distribution and to any small model transport errors.

4 Conclusions

We present the first national-scale top-down emissions estimates of halocarbons for India. We show that India's 2016 halocarbon emissions reflect low emissions of CFCs and regulated chlorocarbons CTC and MCF, and large emissions of HCFCs, HFCs and unregulated chlorocarbons such as DCM. India reported a complete phase-out of its production of CFCs, CTC and MCF by 2010, however banks such as dated refrigeration equipment and insulating foams, as well as fugitive emissions from industry, may persist. Our results indicate that India's remaining major CFC emissions represent 7 (4-12) % of global emissions. Of the refrigerant gases, India's largest emissions are from HFC-134a, HFC-125 and HCFC-22. HFC-134a and HCFC-22 have similar magnitudes of emissions, suggesting that India is in transition between employing HCFC and HFC refrigerants. We present evidence to suggest that India is yet to adopt several common refrigerant blends, including R-410A, R-404A and R-507A, all of which are used extensively in the developed world. India's apparent lack of uptake of refrigerant blends presents an opportunity for future climate mitigation strategies; if India can be encouraged to bypass HFCs in favour of low-GWP alternatives, substantial CO_2eq emissions could be avoided. We also show that following discontinuation of funding from the CDM, some or all of India's manufacturers of HCFC-22 likely resumed venting of the HFC-23 by-product.

Our results indicate that small sources of MCF remain in India and we present evidence that India's CTC emissions are likely a by-product of chloromethane (DCM and chloroform) manufacture. Interest in the global emissions of unregulated chlorocarbons such as DCM, chloroform and PCE has grown in recent years, as increasing emissions from Asia pose a potential threat to the recovery of the ozone layer. Our DCM emissions estimate suggests a 5-fold increase in India's emissions since 2008.

As India's economy expands, its production and consumption of halocarbons is likely to increase dramatically. It is important to implement long-term and continuous halocarbon monitoring from this region of the world to help India evaluate its progress under the Montreal Protocol. Our 2016 estimates provide a benchmark, against which future changes to India's halocarbon emissions can be assessed.

5 Data availability

Data are available from the Centre for Environmental Data Analysis (CEDA):
http://catalogue.ceda.ac.uk/uuid/e838a628dacc438ab4749b011ae7225f

6 Code availability

Hierarchical Bayesian trans-dimensional MCMC code is available on request from Anita Ganesan (Anita.Ganesan@bristol.ac.uk).

Author contributions. D.S. collected the samples, conducted the instrumental analyses, ran the inverse model and wrote the paper. A.G. coordinated the flight campaign, co-developed the inverse model code and aided in the writing of the paper. M.L. co-developed the inverse modelling code and conducted the MOZART model runs. M.R. aided the inverse modelling work. S. O'D. co-conducted the instrumental analyses and provided data from Mace Head. C.H. produced the SIO calibration scales for these gases. A.M. advised on NAME modelling. P.K. provided measurements from Cape Grim. S.B. co-collected the whole air samples.

Competing interests. The authors declare that they have no conflict of interest.

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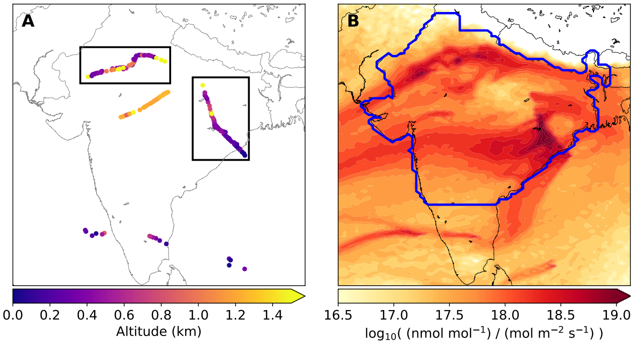

Figure 1. (A) Location and altitude of aircraft samples collected over India. The flight paths outlined in boxes were repeated three times each over the sampling period. (B) Average sensitivity to surface emissions from all samples collected over India. A region broadly corresponding to maximum sensitivity in the samples is shown in the blue outline. We denote this region as northern-central India (NCI). The full inversion domain is shown in Fig. S1.

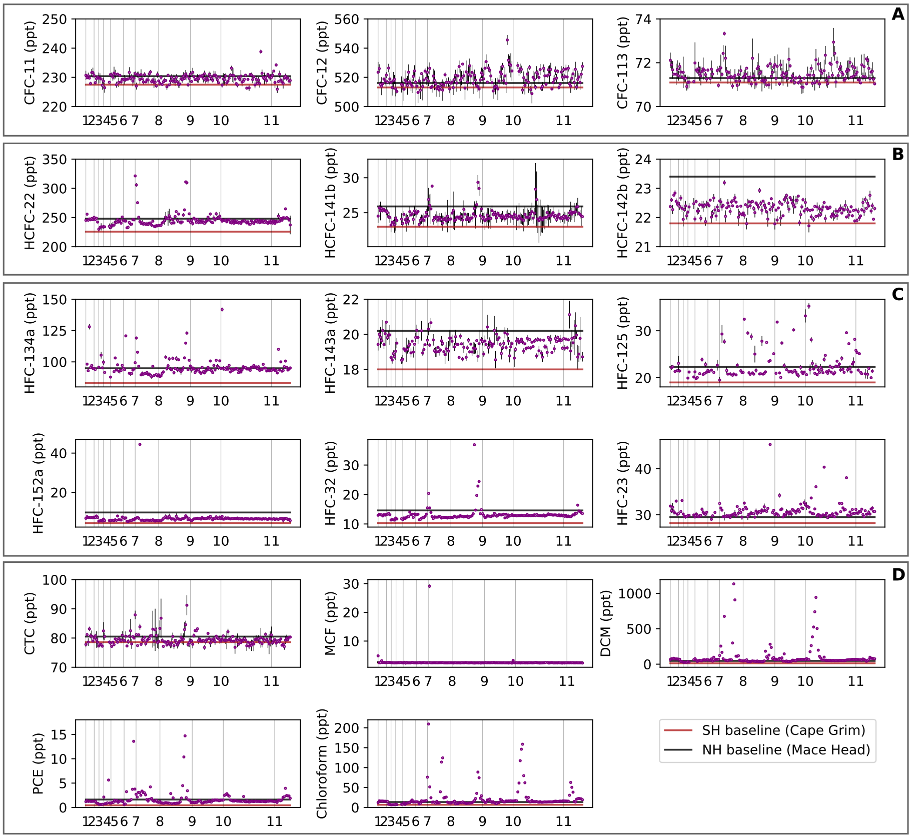

Figure 2. (A) CFC, (B) HCFC, (C) HFC and (D) chlorocarbon mole fraction data from 176 flask samples collected over India, plotted on a flight by flight basis (a summary of flights 1-11 is given in Table 2). Error bars represent instrumental precision, which was estimated using the standard deviation of the three replicate analyses of each flask. Two statistical baselines, inferred from observations at Cape Grim, Tasmania (red line) in the Southern Hemisphere (SH) and Mace Head, Ireland (black line) in the Northern Hemisphere (NH), are shown for comparison.

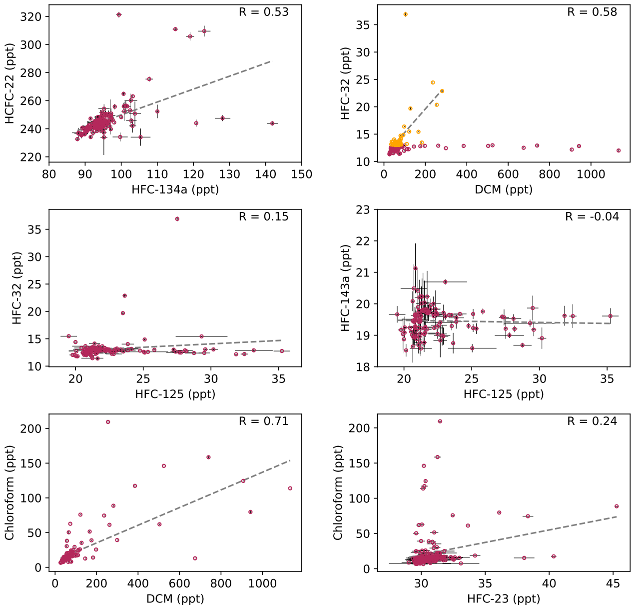

Figure 3. Halocarbon scatter plots, shown with line of best fit and Pearson (R) correlation coefficient. For HFC-32 versus DCM, the sub-scatter shown in orange is a subset of the dataset corresponding to samples whose HFC-32 mole fraction (lower bound of measurement uncertainty) exceeded the 20^{th} percentile of all measurements (and were hence classified as enhanced), since there are likely other sources of DCM not linked to HFC-32 production.

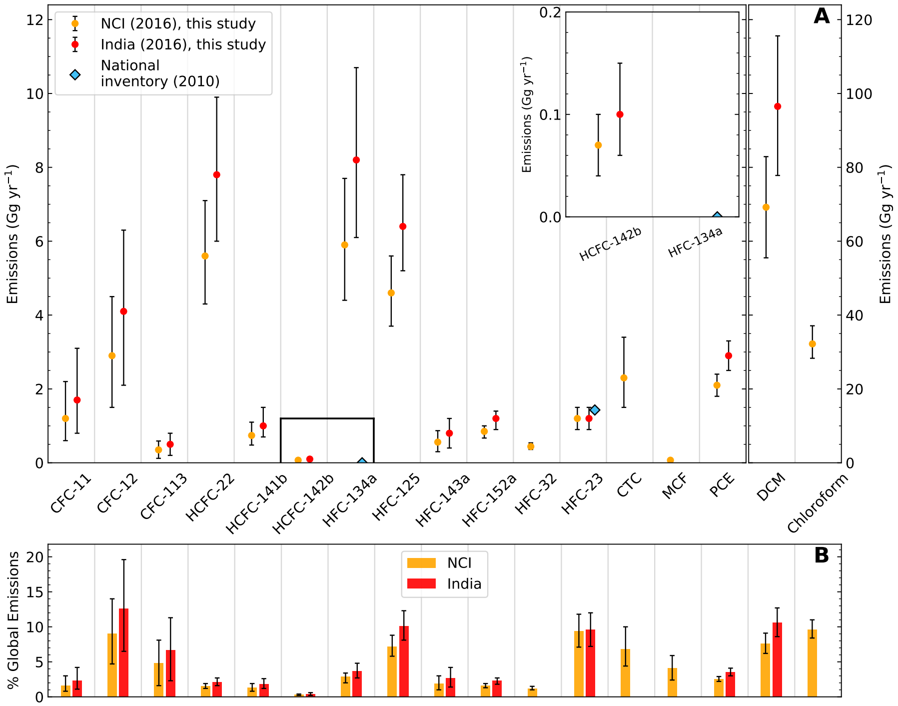

Figure 4. (A) NCI (orange) and India total (red) halocarbon emissions (Gg yr^{-1}) derived in this study. India's most recent greenhouse gas inventory estimates (2010) are included where available. Note that emissions of DCM and chloroform are presented on a second y-axis for clarity. (B) The estimated contribution of the NCI and India to global halocarbon emissions (global estimates are an extension of the work by Rigby et al. (2014)). Error bars represent the 5^{th}-95^{th} percentiles of the posterior distribution.

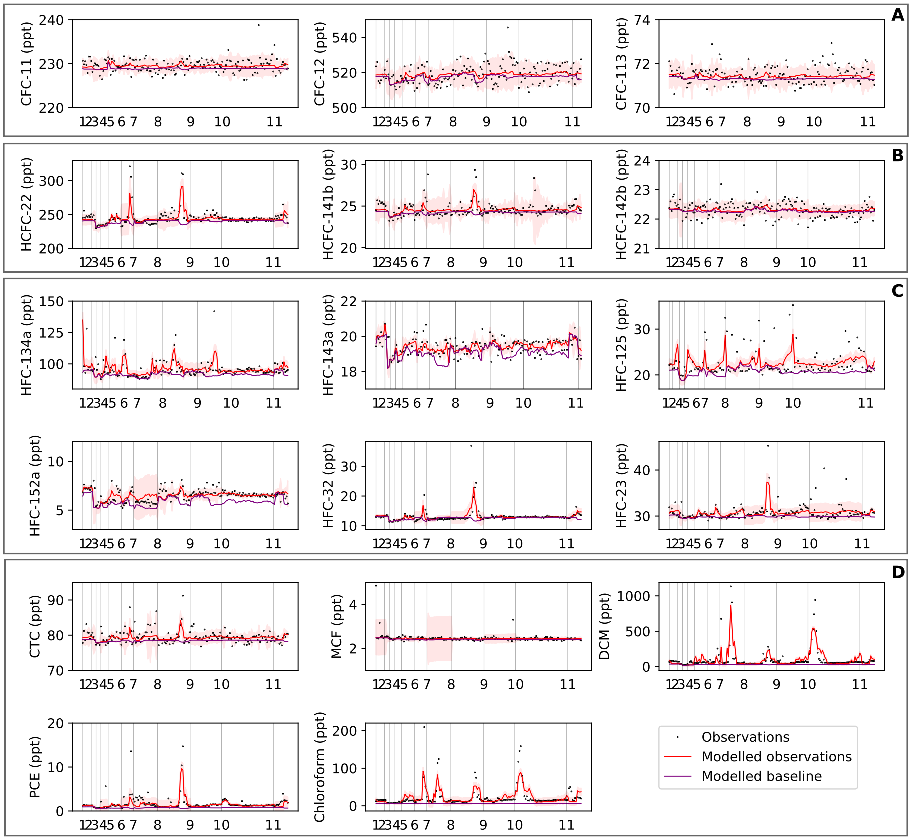

Figure 5. Comparison of measured (black points) with posterior modelled (red line) halocarbon mole fraction data, plotted on a flight by flight basis (a summary of flights 1-11 is given in Table 2). The posterior modelled baseline is also shown (purple line). The shading represents the model uncertainty ($5^{th} - 95^{th}$ percentile of the posterior PDF). With the exception of chloroform, for which prior emissions were distributed uniformly over ocean and land, prior emissions were distributed according to the NOAA night light distribution. Note that for HFC-152a and MCF the y-axis has been reduced in comparison to Fig. 2 for clarity.

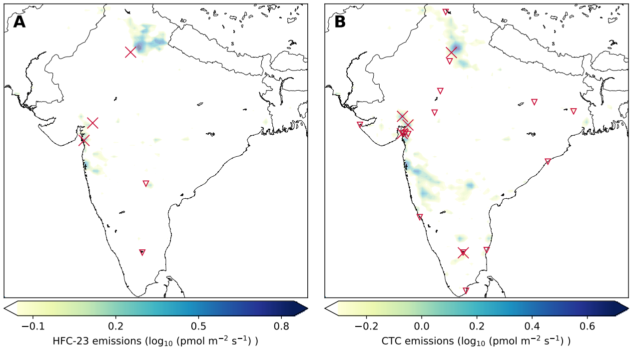

Figure 6. (A) Posterior emissions map for HFC-23, reported in pmol m^{-2} s^{-1}. The known locations of major (> 8 Gg yr^{-1}) and minor (< 1.5 Gg yr^{-1}) manufacturers of HCFC-22 are represented by the crosses and open triangles respectively. (B) Posterior emissions map for CTC, reported in pmol m^{-2} s^{-1}. The known locations of chloromethane production facilities (crosses) and chlor-alkali plants (open triangles) are also shown.

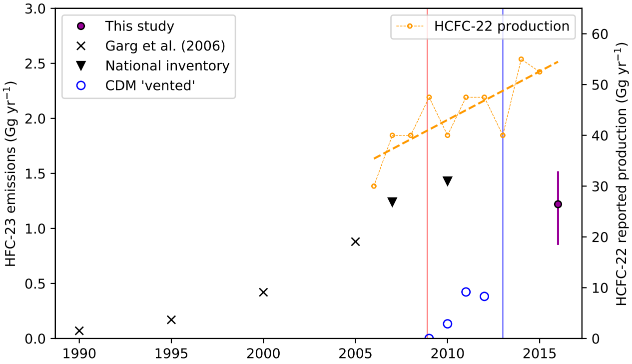

Figure 7. HFC-23 emissions (Gg yr^{-1}) from bottom-up and top-down estimates. Bottom-up estimates are from Garg et al. (2006) (black crosses) and India's Second National Communication Ministry of Environment, Forest and Climate Change (2012) and Biennial Update Report Ministry of Environment, Forest and Climate Change (2015) to the UNFCCC for 2007 and 2010, respectively (black triangles). The top-down estimate derived here is plotted as a purple circle with corresponding 5^{th}-95^{th} percentile uncertainties. Blue circles show the total amount of 'vented' (i.e. released to the atmosphere) HFC-23 per year, as reported by the five HCFC-22 manufacturers during the CDM period. Reported HCFC-22 production (Gg yr^{-1}) data is shown in orange circles and extrapolated to 2016 using a linear fit (dashed orange line). The red bar indicates the first year (2009) in which all five manufacturers of HCFC-22 reported the use of an abatement system and the blue bar indicates the point (January 2013) at which the European Union banned the use of HFC-23 credits under the EU Emissions Trading Scheme. Note the split y-axes – HFC-23 emissions estimates are plotted with respect to the left-hand axis, while HCFC-22 production data is plotted with respect to the right-hand axis.

Table 1. Halocarbons considered in this study. Atmospheric lifetime estimates, ozone-depletion potentials (ODPs) and global warming potentials (100-year time horizon, GWP_{100}) are taken from the 2018 Scientific Assessment on Ozone Depletion (Engel et al., 2019). Lifetimes are quoted in years unless otherwise stated.

Species	Formula	Lifetime	ODP	GWP_{100}	Main application
CFC-11	CCl_3F	52	1.00	5160	Refrigerant
CFC-12	CCl_2F_2	102	0.77	10300	Refrigerant
CFC-113	CCl_2FCClF_2	93	0.81	6080	Solvent
HCFC-22	$CHClF_2$	11.9	0.029	1780	Refrigerant
HCFC-141b	CH_3CCl_2F	9.4	0.086	800	Foam-blowing
HCFC-142b	CH_3CClF_2	18	0.040	2070	Foam-blowing
HFC-134a	CH_2FCF_3	14	0	1360	Refrigerant
HFC-143a	CH_3CF_3	51	0	5080	Refrigerant
HFC-125	CHF_2CF_3	30	0	3450	Refrigerant
HFC-152a	CH_3CHF_2	1.6	0	148	Aerosol propellant
HFC-32	CH_2F_2	5.4	0	705	Refrigerant
HFC-23	CHF_3	228	0	12690	By-product
CTC	CCl_4	32	0.89	2110	Cleaning agent
MCF	CH_3CCl_3	5.0	0.155	153	Cleaning agent, degreaser
DCM	CH_2Cl_2	180 days	Not well quantified	10	Solvent, feedstock
PCE	C_2Cl_4	110 days	Not well quantified	5.9	Dry cleaning agent
Chloroform	$CHCl_3$	183 days	Not well quantified	18	Feedstock

Table 2. Aircraft campaign flight summary statistics. IST – Indian Standard Time

Flight number (Fig. 2 label)	Date (time, IST)	Sampling region	Mean altitude (range, km)	Number of samples
B957 (1)	12/06 (06:02 – 07:55)	NE India	1.20 (0.30 – 7.40)	9
B959 (2)	21/06 (08:10 – 08:21)	S India	0.46 (0.05 – 0.87)	2
B963 (3)	25/06 (16:52 – 18:00)	S India	0.31 (0.21 – 0.53)	4
B966 (4)	27/06 (07:12 – 09:49)	S India	0.30 (0.02 – 0.66)	9
B968 (5)	30/06 (05:03 – 06:51)	NW India	0.98 (0.28 – 3.15)	11
B969 (6)	02/07 (05:21 – 07:11)	NW India	0.53 (0.28 – 0.64)	11
B971 (7)	04/07 (07:23 – 08:57)	NE India	0.38 (0.02 – 1.65)	20
B972 (8)	05/07 (05:23 – 07:06)	NW India	0.83 (0.30 – 1.65)	27
B974 (9)	07/07 (06:22 – 07:30)	NW India	1.29 (0.88 – 2.90)	26
B975 (10)	09/07 (06:31 – 08:14)	NE India	0.37 (0.02 – 1.16)	44
B976 (11)	10/07 (06:37 – 07:32)	NW India	0.42 (0.35 – 0.53)	12

Table 3. Halocarbon mass spectrometry target/qualifier ions and respective calibration scales. SIO - Scripps Institution of Oceanography, NOAA - National Oceanic and Atmospheric Administration. Average flask measurement precisions are also shown.

Species	Target ion (*m/z*)	Qualifier ion (*m/z*)	Calibration scale	Average measurement precision (%)
CFC-11	103	105	SIO-05	0.4
CFC-12	85	87	SIO-05	0.7
CFC-113	153	155	SIO-05	0.4
HCFC-22	67	50	SIO-05	0.9
HCFC-141b	81	101	SIO-05	3.4
HCFC-142b	65	85	SIO-05	0.5
HFC-134a	83	33	SIO-05	0.8
HFC-143a	65	64	SIO-07	1.3
HFC-125	101	51	SIO-15	1.5
HFC-152a	65	46	SIO-05	3.2
HFC-32	33	51	SIO-07	1.1
HFC-23	51	69	SIO-07	0.8
CTC	82	84	SIO-05	1.3
MCF	99	97	SIO-05	1.7
DCM	86	84	SIO-14	1.0
PCE	166	164	NOAA-2003B	1.0
Chloroform	83	85	SIO-98	0.5

Table 4. Posterior mean halocarbon emission estimates reported in Gg yr^{-1} for northern-central India and the whole of India, and the percentage contribution of India to global emissions, where appropriate. The 5^{th} and 95^{th} percentile ranges are shown in parentheses. Asterisks denote that percentages are derived from the NCI total, as scaling to a national total was not considered appropriate for these gases.

Species	NCI Prior	NCI Posterior	India	% of global
CFC-11	9.0	1.2 (0.6 – 2.2)	1.7 (0.8 – 3.1)	2.3 (1.1 – 4.2)
CFC-12	4.1	2.9 (1.5 – 4.5)	4.1 (2.1 – 6.3)	12.6 (6.5 – 19.6)
CFC-113	0.89	0.35 (0.12 – 0.59)	0.49 (0.17 – 0.82)	6.7 (2.3 – 11.3)
HCFC-22	8.0	5.6 (4.3 – 7.1)	7.8 (6.0 – 9.9)	2.1 (1.6 – 2.7)
HCFC-141b	2.1	0.7 (0.5 – 1.1)	1.0 (0.7 – 1.5)	1.8 (1.2 – 2.6)
HCFC-142b	0.09	0.07 (0.04 – 0.10)	0.10 (0.06 – 0.14)	0.4 (0.2 – 0.6)
HFC-134a	5.9	5.9 (4.4 – 7.7)	8.2 (6.1 – 10.7)	3.7 (2.7 – 4.8)
HFC-143a	1.4	0.56 (0.30 – 0.87)	0.8 (0.4 – 1.2)	2.7 (1.4 – 4.2)
HFC-125	1.5	4.6 (3.7 – 5.6)	6.4 (5.2 – 7.8)	10.1 (8.1 – 12.3)
HFC-152a	1.1	0.9 (0.7 – 1.0)	1.2 (0.9 – 1.4)	2.3 (1.8 – 2.7)
HFC-32	0.11	0.44 (0.36 – 0.54)	-	1.2 (1.0 - 1.5)*
HFC-23	1.1	1.2 (0.9 – 1.5)	1.2 (0.9 – 1.5)	9.6 (7.2 – 12.0)
CTC	2.0	2.3 (1.5 - 3.4)	-	6.8 (4.4 - 10.0)*
MCF	0.2	0.07 (0.04 - 0.10)	-	4.1 (2.4 - 5.9)*
DCM	14.6	69.2 (55.5 - 82.9)	96.5 (77.8 - 115.6)	10.6 (8.6 - 12.7)
PCE	4.3	2.1 (1.8 - 2.4)	2.9 (2.5 - 3.3)	3.5 (3.0 - 4.1)
Chloroform	2.2	32.2 (28.3 - 37.1)	-	9.6 (8.4 - 11.0)*

Table 5. Posterior mean halocarbon emission estimates reported in Tg CO_2eq yr^{-1} for northern-central India and the whole of India. The 5^{th} and 95^{th} percentile ranges are shown in parentheses. Emissions totals for the whole of India are not presented for HFC-32, CTC, MCF and chloroform, as scaling to a national total was not considered appropriate for these gases.

Species	NCI Prior	NCI Posterior	India
CFC-11	46.4	6.2 (3.1 – 11.4)	8.8 (4.1 – 16.0)
CFC-12	42.2	29.9 (15.5 - 46.4)	42.2 (21.6 - 64.9)
CFC-113	5.4	2.1 (0.7 - 3.6)	3.0 (1.0 - 5.0)
HCFC-22	14.2	10.0 (7.7 - 12.6)	13.9 (10.7 - 17.6)
HCFC-141b	1.7	0.6 (0.4 - 0.9)	0.8 (0.6 - 1.2)
HCFC-142b	0.19	0.14 (0.08 - 0.21)	0.21 (0.12 - 0.29)
HFC-134a	8.0	8.0 (6.0 - 10.5)	11.2 (8.3 - 14.6)
HFC-143a	7.1	2.8 (1.5 - 4.4)	4.1 (2.0 - 6.1)
HFC-125	5.2	15.9 (12.8 - 19.3)	22.1 (17.9 - 26.9)
HFC-152a	0.16	0.13 (0.10 – 0.15)	0.18 (0.13 – 0.22)
HFC-32	0.08	0.31 (0.25 – 0.38)	-
HFC-23	14.0	15.2 (11.4 – 19.0)	15.2 (11.4 – 19.0)
CTC	4.2	4.9 (3.2 - 7.2)	-
MCF	0.03	0.01 (0.01 - 0.02)	-
DCM	0.15	0.69 (0.56 - 0.83)	0.97 (0.78 - 1.1)
PCE	0.03	0.01 (0.01 - 0.01)	0.02 (0.01 - 0.20)
Chloroform	0.04	0.58 (0.51 - 0.67)	-

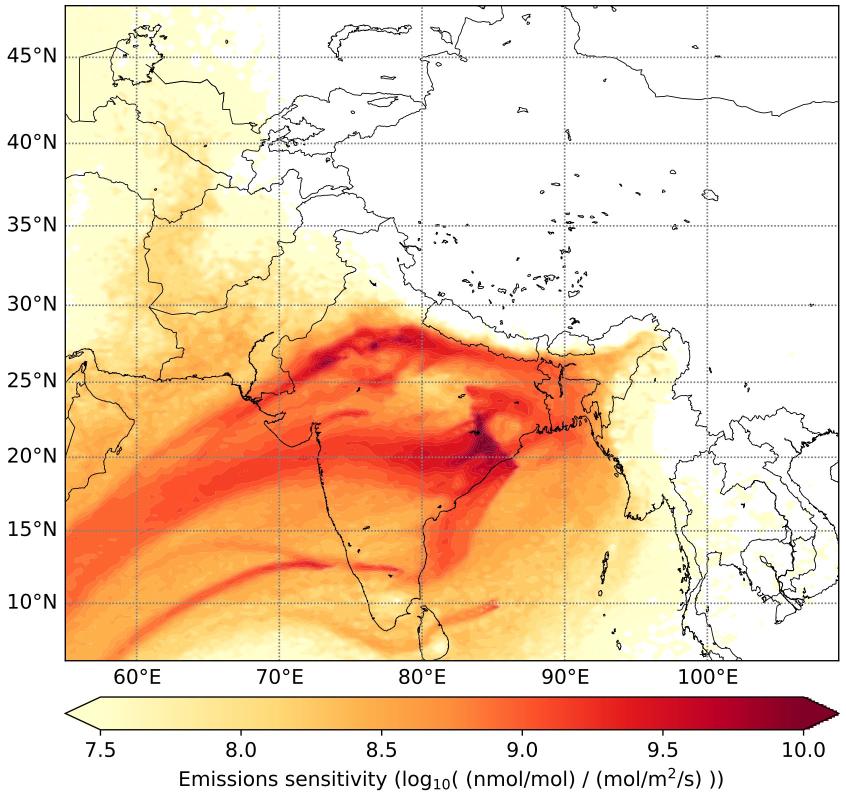

Figure S1. Average sensitivity to surface emissions from all samples collected over India across the entire NAME model domain.

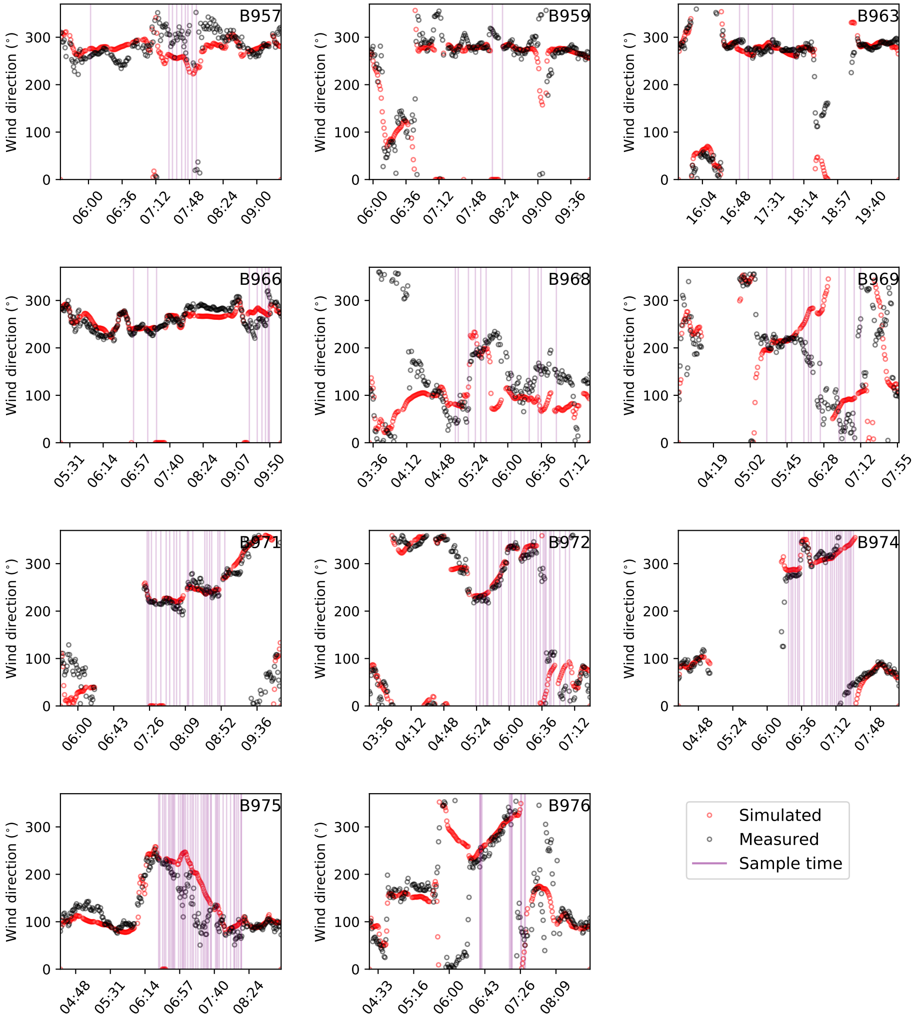

Figure S2. Comparison of wind direction (°), as measured from the research aircraft (black circles) with those simulated by the NAME model (red circles), for each flight on a minute by minute basis. The points in time at which samples were collected are indicated by vertical purple lines.

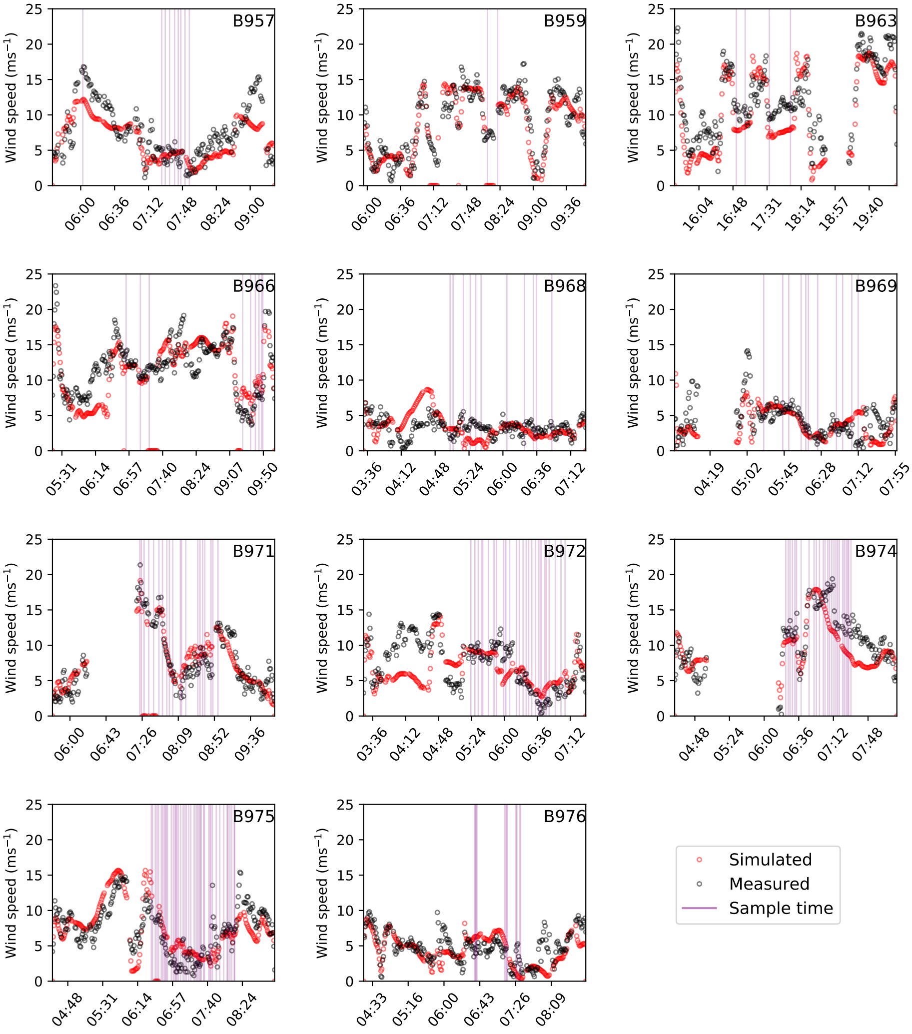

Figure S3. Comparison of wind speed (m s^{-1}), as measured from the research aircraft (black circles) with those simulated by the NAME model (red circles), for each flight on a minute by minute basis. The points in time at which samples were collected are indicated by vertical purple lines.

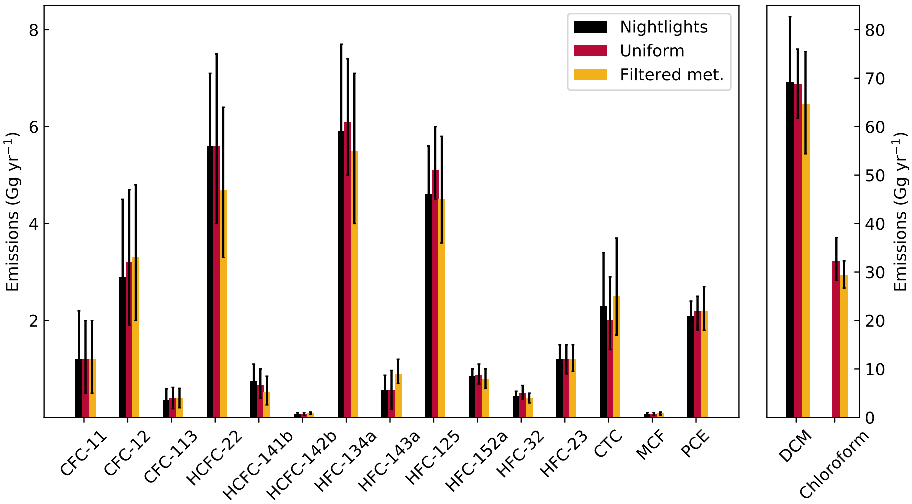

Figure S4. Comparison of posterior NCI halocarbon emissions estimates. Black bars are based on prior emissions distributed using the NOAA night lights distribution; red bars are based on prior emissions distributed using uniformly across the model domain; yellow bars are based on the NOAA night lights distribution, but has removed observations where the observed and model simulated wind speed/direction differed by more than 20% removed. Error bars represent the $5^{th} - 95^{th}$ percentiles of the posterior PDF. Note that DCM and chloroform are presented on their own y-axis for clarity.