Many thanks to both the reviewers for their constructive comments. Our responses and actions are given in detail below (blue text).

Referee 1

Comments on: Evidence of a recent decline in UK emissions of HFCs determined by the InTEM inverse model and atmospheric measurements, Manning et al., 2021, https://doi.org/10.5194/acp-2021-261

This paper describes results from an inverse model study of HFC emissions from the UK using the Numerical Atmospheric dispersion Modelling Environment (NAME) model incorporated into the Inversion Technique for Emissions Modelling (InTEM) model system. This study uses data from long-term monitoring sites at Mace Head, Ireland; Jungfraujoch, Switzerland; and Monte Cimone, Italy. In addition, the study incorporates recent measurements from Tacolneston (UK), Carnsore Point (Ireland), and Taunus (Germany) to better constrain UK emissions and provide higher spatial resolution. The authors found that total UK emissions of the HFCs studied were lower in 2019-2020 (expressed in CO2-equivalent units) than in the previous decade. They also found that UK emissions based on top-down methods are about 30-50% lower than bottom-up, inventory-based emissions for several HFCs.

This work and the underlying methods provide important constraints and feedback on policy decisions related to reducing the consumption of HFCs in order to limit the climate impacts of these high-global-warming potential gases. This work should also help inform and possibly improve bottom-up inventory estimates of these gases.

General Comments:

The paper is well-written and the methods are well-established. I have only a few comments related to the work and presentation.

1. On page 4, line 94, you say that you are applying an updated version of the InTEM model framework. Since models are updated occasionally, it would help the reader to know if the version of InTEM used in this study is the same as that described in Arnold et al (2018). If updates have been made since Arnold et al (2018), perhaps these could be summarized somewhere.

   This is a good point, we have reviewed all model changes since Arnold (2018) and added additional text to the paper and a bullet point summary into the Supplementary Material.

   Text added on line 149:

   'A summary of InTEM model changes from Arnold et al 2018 to the current InTEM model are provided in the Supplementary Material.'

   Some re-wording for greater clarity has been included, line 219:

   'There are 11 boundaries encompassing the computational domain which are adjusted independently. They are described in detail in Fig. 2 of Arnold et al (2018), however note that the lower boundary in the current study is now 3 km above ground and the upper boundary is now 3--8 km.'

Additional text in the SM:

'InTEM has been improved since it was reported in detail in Arnold et al 2018. The significant updates are listed here:

- The prior estimate of the background has been refined further. A daily, rather than hourly, background value is calculated after selecting those 2-hourly windows classified as background by fitting a 4th-order, 2nd-order or 1st-order (previously just 2nd-order) polynomial depending on the number of background points in a moving 6-month window (40-150 days previously).
- Time-integrated mole fractions (mol mol^{-1} s) are output from NAME which internally used the modelled temperature and pressure at the output location. Previously time-integrated air concentrations (g s m^{-3}) were output and subsequently converted using the station temperature and pressure. The latter is far less accurate especially when pertaining to high altitude stations.
- NAME was run using 20,000 rather than 5,000 particles per hour to reduce computational noise.
- InTEM now solves for a bias between the different observation stations, applicable here as different observation stations are used.
- The model uncertainty applied to each model time-period (4-hours in this study) has been significantly revised and is detailed in the main text.
- The heights of the 11 directions related to where the air enters the computational domain have been modified from 0--6 km, 6--9 km and >9 km to 0--3 km, 3--8 km and >8 km, relating to boundary layer/lower tropospheric, free-tropospheric and upper-tropospheric respectively.
- Each inversion undergoes 24 'zooming' steps rather than 50, the extra steps were found to be irrelevant in the UK context.'

2. The method of running the inversion in 1-yr and 2-yr blocks could use a bit more explanation. Why is this done? Is this new or typical?

Thank you for this comment, we have added a sentence to the paper as follows:

'2-year inversion periods are necessary pre-2013 to increase the amount of data that the inversion system has to constrain UK posterior emission estimates. 1-year inversion UK estimates with TAC data give a more realistic, and thus smoother, year-to-year variability in the UK emissions; see Supplementary Material for further information and Table 2 for details of data availability.'

Also we have added a section to the Supplementary Material as follows:

'Figure S3 shows the 1 and 2-year inversion estimates for HFC-134a from 1994. The purpose of this figure is to demonstrate the unrealistic year to year variability in the 1-year inversion data when there is only one data site operating (MHD). From 2003 data are available from JFJ and

CMN, from CSP between 2005 and 2010, and from TAC and TOB from 2013. 2-year inversion periods are necessary initially to increase the amount of data that the inversion system has to constrain the posterior emission estimate. 1-year inversion estimates using only data from MHD gave an unrealistic year to year variability in the UK emissions. There is closer agreement in the 1-year and 2-year inversions as more data becomes available and hence the ability to present a 1-year inversion emission estimate for the UK from 2013'

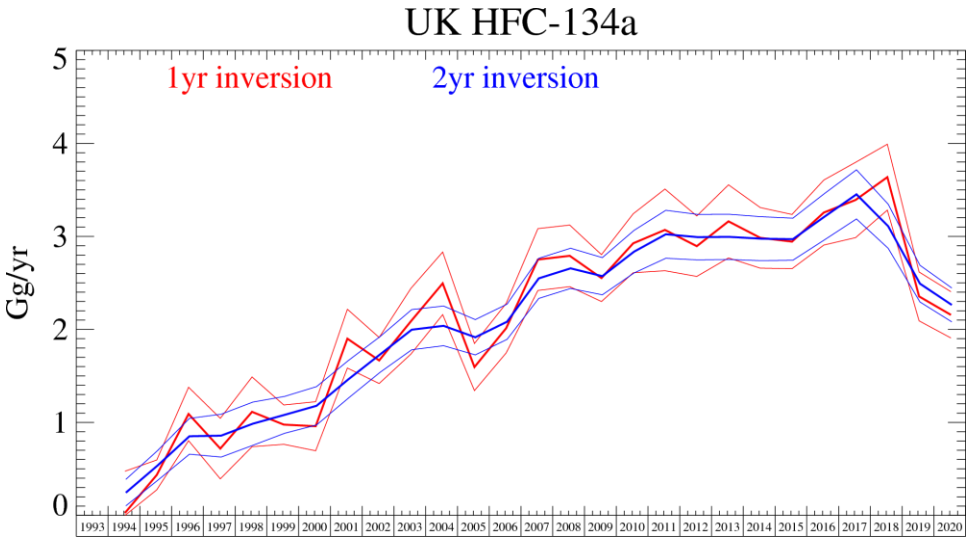


Figure S3: Annual UK InTEM emission estimates (Gg), annualised 2-year inversion (blue) and 1-year inversion (red) for HFC-134a. The uncertainty bars represent 1σ.

3. On page 7, line 159, you discuss the use of a uniform prior emission field for HFC-23, and then later explain that you use 100% uncertainty on UK emissions in the prior for other gases. I would assume that a uniform (flat) prior would also lead to similar results for other gases (little dependence on the prior). Perhaps you can comment if that is indeed the case.

   Thank you for this comment, additional sensitivity InTEM inversions were performed to demonstrate the lack of sensitivity to the prior. Additional text was added to the end of the HFC-134a results section in the main paper.

   'As a sensitivity test, the HFC-134a 1-year inversions were also performed using a uniform land-based (flat) prior. The InTEM 2013--2020 average UK emissions were similar when using the flat prior (4.1 +- 0.45 Tg CO_2-eq) compared to when using the population prior (4.1 +- 0.54 Tg CO_2-eq), showing that there is little dependence on the prior.'

4. In section 2.3 you refer to both "background" and "baseline" mole fractions. Are these the same thing? Or is "background" a model term and "baseline" a measurement term? It is a minor point, but these terms seem to be used interchangeably. And you also refer to background mole fractions as "the prior" (pg. 6, line 145). Not to be confused with the emissions "prior". Is there a better way to distinguish between these different things?

Thank you for pointing out this inconsistency. The use of baseline has been removed throughout the paper and replaced with background. Text giving a definition of what we mean by background in the context of the paper is given below and has been included in the inversion framework section.

'In the context of this paper, the background is defined as the mole fraction of the gas as it enters into the defined computational domain, this will be representative of the well-mixed Northern Hemisphere mole fraction adjusted based on the direction and height of the air entering the domain'

Specific Comments:

P 8, L194: "Time-varying background levels of mole fractions are required as prior information for InTEM for three stations: MHD, JFJ and CMN."

This implies that background mole fractions are not needed for the other stations (TAC, CSP, TOB). Is that the message you intended?

Thank you for identifying this mistake. A background is required at these 3 stations, we use the MHD background. This has now been made clear in the text in the inversion framework section.

'The prior background mole fractions from MHD are used at CSP, TAC and TOB. However, it is important to note that as the air reaching these stations enters the computational domain from different directions, the prior and posterior background mole fractions at each station will be different from MHD and each other.'

P 17, L280: " …however the growth in the Northern Hemisphere appears to have peaked in 2017 …"

Given that the background mole fraction HFC-134a shows interannual variability, one could have also said the same thing in ~2006 and ~2013. So perhaps it is too soon to say? Still, I like figure 4 because it provides global context.

This is a good point and the following text has been added to this paragraph to caveat the statement.

'It should be noted, as Fig 4 shows, there is evidence of previous inter-annual variability in the growth of HFC-134a, so further years of data will be required to confirm this.'

Sec. 3.2, HFC-134a: Perhaps a comment on COVID-19 is also applicable here. Is there any relation between HFC-134a emissions and vehicle miles driven? Could the lock-downs have played a role in 2020? Perhaps not, since 2019 emissions were also lower. It might also be relevant to mention when the EU began the transition to HFO alternatives to HFC-134a in mobile air conditioning.

We have added a comment to this effect in Section 3.2:

'It is possible that there was a change in UK HFC-134a emissions due to the Covid-19 pandemic in 2020, however the most significant drop in UK emissions occurred in 2019, before the pandemic arrived in the

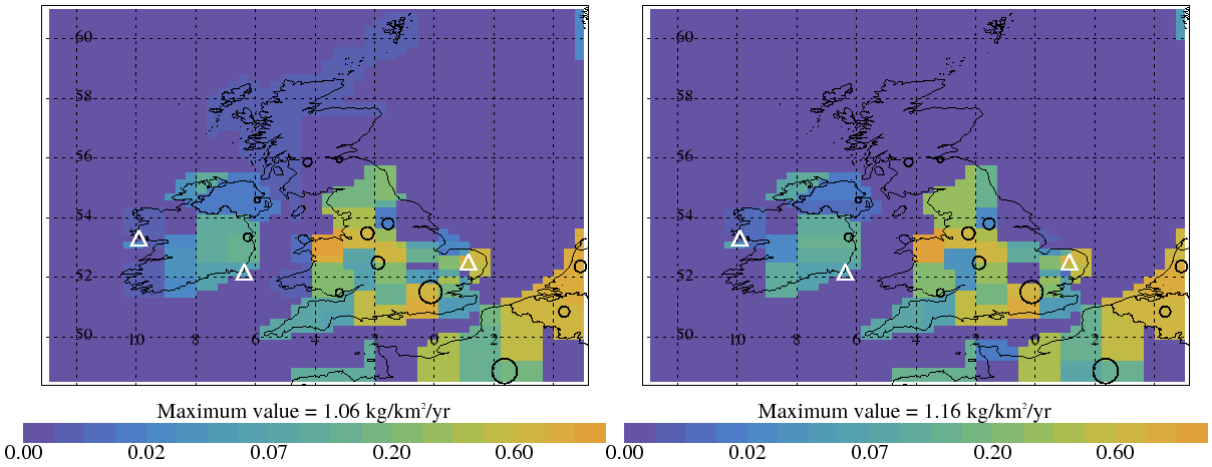
UK. It is probable therefore that the Covid-19 impact on UK HFC-134a emissions, if any, was small, but ultimately cannot be ascertained yet.'

Figure 6: The emission source maps for most HFCs shown seem closely related to population density, except for HFC-227ea, which shows a region of emissions in NW England that does not correspond to a large population center. And the correlation coefficients in Table S2 are smaller for some observing sites. Is this worth a comment, especially with respect to results from different years? Is that region of emissions in NW England an anomaly in 2019-2020?

Thank you for this comment, we have changed the text to indicate that these features are seen across the years:

'The spatial pattern of the InTEM emission estimates are shown in Fig. 6 and they largely follow population, though with more variation in the spatial distribution than is seen in the other HFCs. This effect continues across the years, with high emissions in NE Wales (low population) and NW England (high population) appearing from 2015; high emissions are also observed from the near continent. It is also notable that the statistical fit between the observed and modelled time-series in all years, principally at TAC (see Supplementary Material for more details), is significantly worse than for the other gases. The uses of HFC-227ea, for example, as the UK's dominant HFC fire-suppression gas, means that its emission is potentially much more intermittent, thus making it harder to model. It is also likely to be less tied to population distribution because of its specialist uses. Similarly to HFC-134a, a uniform land-based (flat) prior was tested, the results show remarkable similarity (average 2013-2020 UK emissions: 0.19 +/- 0.039 Tg CO_2-eq) to the results using the population prior (average 2013-2020 UK emissions: 0.18 +/- 0.033 Tg CO_2-eq).'

The comparison between using a uniform land (flat) and a population prior is shown below, the maps are essentially identical. This shows the prior has negligible impact on the spatial or UK emission totals.



Technical Corrections:

P, L15: remove "in the" after "decline in UK emissions". "… decline in UK emissions in the since 2018".

Done

Referee 2

Comments on: Evidence of a recent decline in UK emissions of HFCs determined by the InTEM inverse model and atmospheric measurements, Manning et al., 2021, https://doi.org/10.5194/acp-2021-261

The manuscript assesses the emissions rate of HFCs from the UK through a top down approach. The inversion method is based on the Numerical Atmospheric dispersion Modelling Environment (NAME) model in conjunction with measurement observations and the Inversion Technique for Emissions Modelling (InTEM) model system. The inverse modelling results point out a decreasing trend of the aggregated HFCs emissions since 2018, in response to the UK implementation of European Union regulation of those gasses emissions. Moreover, the aggregated HFCs emissions calculated are significantly lower (73%) than those reported from the UK to National Greenhouse Gas Inventories (GHGI) and submitted annually to the United Nations Framework Convention on Climate Change (UNFCCC).

General comments

This work is well written, and it is very much appreciated.

Few general comments:

1. On page 4 line 98, Does the Carnsore Point, Taunus and Tacolneston measurement data have different calibration scales compared to Mace Head and the two mountain stations ? If the authors provided the intercalibration between those scales it could be interesting to show it on Table 2.
   Good point, we have added a sentence about the calibration scales:

   'The measurements of each gas are reported on the same, gas-specific, AGAGE calibration scale irrespective of station.'

2. It could be useful add more information about the Carnsore Point, Taunus and Tacolneston measurement stations (e.g. sub urban, rural or remote).
   Thank you for raising this, these stations are all rural and this has been added to the text.

3. On page 4 line 98, "which greatly enhances the ability…" As you do not show any test that proves it, I would suggest rephrasing it or report some specific test (on Supplementary Material S.M.) that proves how those stations improved the inversion performance.

   Thank you for this important point, we have removed 'greatly' and refer the reader to the SM for details so that the text now reads "which enhance (see Supplementary Material) the ability…".

   Section added to the SM is as follows:
   'S5 Impact of measurement sites on UK HFC-134a emissions estimates

The impact of adding measurement data from additional stations to the inversions on the UK emissions has been assessed by estimating HFC-134a UK totals with and without different measurement data. From 2006–2010 CSP data were included giving data from four sites (MHD, CSP, JFJ and CMN). With CSP data included the 2006–2010 average HFC-134a UK annual total was 3.45±0.29 Tg CO2-eq compared with 3.78±0.34 Tg CO2-eq without. The additional data-set from CSP has lowered the annual average emission estimate for the UK and the standard deviation but the uncertainty estimates still overlap. For the period 2013–2020, there are 5 sites available: MHD, JFJ, CMN, TAC and TOB. Using all data gives an average annual UK emissions estimate (2013–2020) of 4.06±0.45 Tg CO2-eq. Omitting data from TAC gives an estimate of 4.39±0.64 Tg CO2-eq and the removal of data from TOB gives 4.06±0.45 Tg CO2-eq. The effect of the additional data-set from TAC is therefore to lower both the annual average estimate and the standard deviation, but the uncertainties overlap. The addition of the TOB data-set has no impact on the UK estimates. The German station TOB provides only weekly flask data and therefore contributes far less data to the UK estimates than the other sites. The mountain stations (JFJ, CMN) are considerably more distant from the UK and the effect of their impact on the UK annual average 2013–2020 emissions estimate has also been considered. The 2013–2020 UK average without JFJ and CMN is 4.11±0.47 Tg CO2-eq, so again the loss of data from these stations slightly increases the UK estimate and standard deviation. This analysis does not assess the impact of the different data-sets on the spatial distribution of emissions which is much harder to assess quantitatively. The impact of each data-set on any individual annual UK estimate varies depending on data availability at each station. Additional data-sets, even if distant from the UK, allows the surrounding regions to be better constrained thereby improving the UK emission estimate. In all cases more data reduces the uncertainty of the UK estimate.'

4. As the Taunus station is located in the center of Germany, and it has only 1 weekly measurement, does it really greatly improve the ability of the inversion system for the UK?
   This is included in the response to point 3 above.

5. Are the two mountain stations affected by HFCs UK sources? If not, why did you use them on your inversion system?
   This is included in the response to point 3 above.

6. Did you run the inversions for the central Europe (Fig 1) or only for UK and IE (fig 6)? I think showing (in fig 1) the European domain as the inversion model domain, but then reporting emission values only for the UK and Ireland (Fig 6) could generate confusion. Unless, you better explain this approach
   A good point, a sentence has been added to the end of the Introductory section to aid clarity on this question:
   'This work is focused purely on reporting emissions from the UK, future work will expand the emission estimates to a wider European region.'

7. On page 5 line 135 , If I well understand the system, using the passive tracer over 30 days of the simulations, it should imply an underprediction of ~5% of the HFC-152a estimates. If so, you should mention it.

Thank you for this comment, we do not believe this is the case as, rather than 30 days (the maximum model travel time allowed), the impact is related to the travel time from the UK to the observation stations. We have added a sentence to make this point:
'Travel time from the UK to the observation stations is less than 5 days for the vast majority of times for all of the stations, therefore there will be negligible impact due to lifetime on the UK emission estimates even for HFC-152a.'

8. On page 6 line 140, I agree with referee 1, could you please describe the updates on InTem system?
   Thank you, in response we have added a section to the Supplementary Material – please see our response to reviewer 1, comment 1.

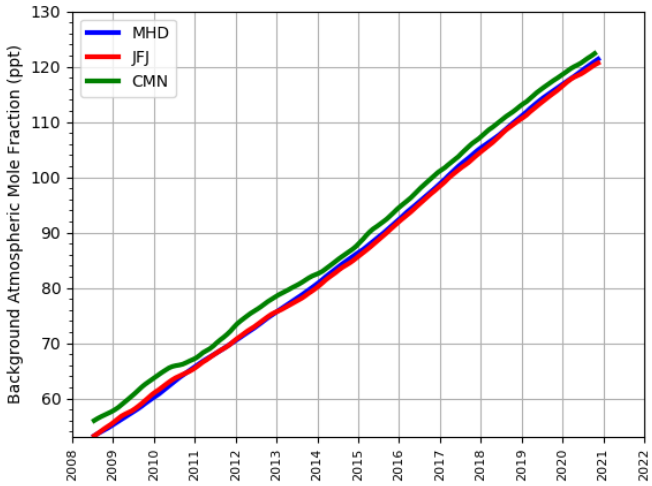
9. On page 9 line 204. "when the population under the surface footprint is small", Did you use the same threshold value used for MHD ?
   An interesting question, thank you. The thresholds used for each station are individually calculated by assessing pollution events relative to each factor at each station. This was done to balance the need for generating sufficient times to allow a good background estimate against the need to exclude polluted times. MHD has many background times so the thresholds can be more strict, whereas CMN has fewer, requiring more lax thresholds to maintain sufficient data for a good background fit.

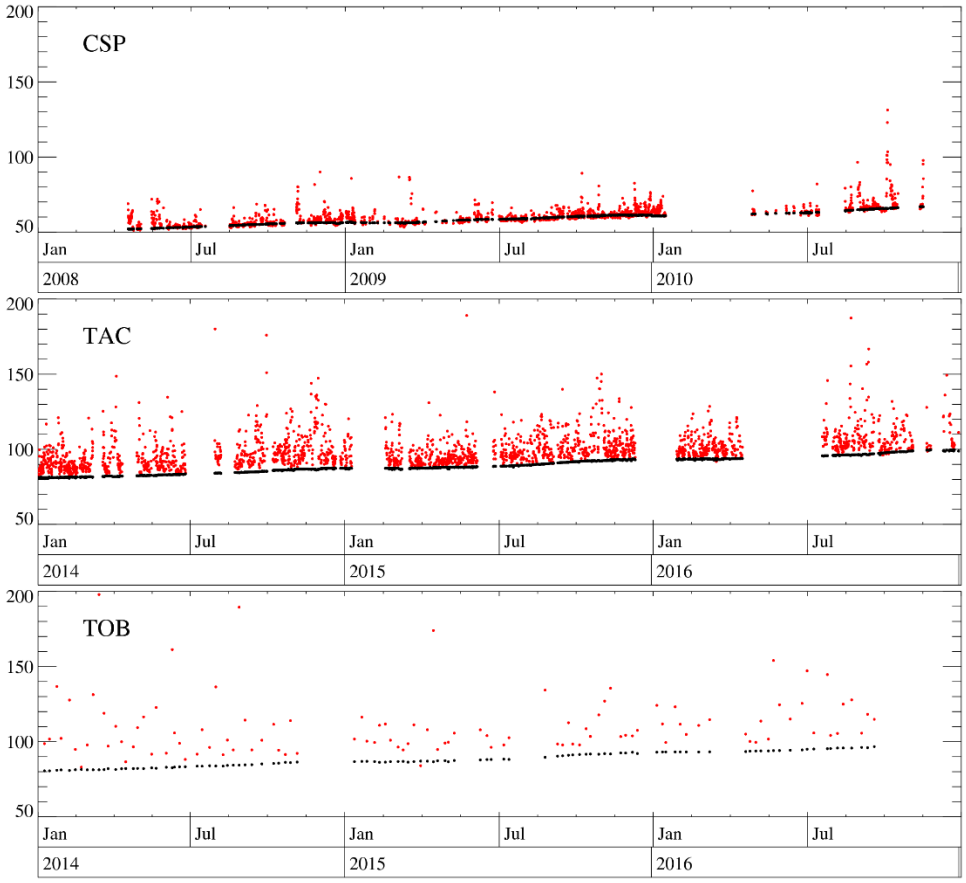
Specific comments:

1. On page 4 line 103, I think you could introduce the Site Acronym, and then use them from this point on.
   Thank you, this has been done.

2. On page 12 paragraph 3.1 I think in this paragraph you should report more quantitative analysis of the main important trends described, instead of qualitative indication .
   Thank you for this comment, but in this case the text was chosen to be qualitative because the quantitative analysis is shown in Figures 3 and 4, the lines are continuous and therefore it is not clear which numbers or averaging times you would choose to add extra value to the comments in the text.

3. On page 12 paragraph 3.1 I think it could be useful to show (on S.M.) the plot of the three baseline data and also the MHD baseline data overlapping the observed time-series of TAC, CSP and TOB.
   Thank you for this suggestion, we have added two additional plots to the Supplementary Material, see below, showing the comparison of the prior background mole fractions for HFC-134a as estimated at the 3 background stations and posterior baseline data against the observed time-series (for 3 year periods for clarity) at TAC, CSP and TOB.





4. On page 18 315 “increase rapidly” please report the value of it.
   Good point, we have inserted actual values relating to the UK estimates into the HFC sections (3.3 – 3.6) to better quantify the written statements.

5. On page 18 334 “accelerating rate” please report the value of acceleration or trend.
   Please refer to answer 2 above as this relates to the growth of the global mole fractions.
6. On page 19 3.6 HFC-227ea “It is also likely to be less tied to population distribution because of its specialist uses” In this case why did you not use a uniform land-based prior as for HFC-23?
   Please refer to the second to last ‘Specific comment’ of reviewer 1.
7. On page 19 348 “HFC-227ea decline markedly from 2018 to 2020” please report the value to indicate it
   Good point, we have added values to the text:
   ‘Similar to the other HFCs reported, InTEM emission estimates of HFC-227ea decline markedly from 2018 (0.25 Tg CO_2-eq) to 2020 (0.16 Tg CO_2-eq).’

8. I agree with referee 1, a few times the authors use “background” but it seems they rather refer to “baseline”. See below the difference and the definition of both words: “Baseline concentrations refer to observations made at a site when it is not influenced by recent, locally emitted or produced anthropogenic pollution. The term global or hemispheric background concentration is a model construct that estimates the atmospheric concentration of a pollutant due to natural sources only.” References: HTAP, T., 2010. Hemispheric Transport of Air Pollution 2010 Part A: Ozone And Particulate Matter, Air Pollution Studies No. 17. Cooper, O. R., Parrish, D. D., Ziemke, J., Cupeiro, M., Galbally, I. E., Gilge, S., ... & Oltmans, S. J. (2014).

   Thank you for your suggestions – please see the response to point 4 from reviewer 1.

9. On page 2 Table 2 of Supplementary Material. Do the 1 yr inversion correlation values are similar to those ? For clarity, if so, just mention it on the paper or report the Table of 1 yr inversion correlation values.
   Thank you for this suggestion, we have added a sentence to this section of the Supplementary Material:
   ‘The 1-yr InTEM statistics are very much aligned to these values, for example the average 2019 and 2020 HFC-134a correlation coefficients for MHD, TAC, JFJ, CMN and TOB are 0.96, 0.80, 0.83, 0.71 and 0.43 respectively.’

10. On page 6 line 19 of S. M. . “ a significant decline “ please report a value of significant decline
    Thank you for pointing this out, we have inserted actual values relating to the UK estimates into the HFC sections (SM 8.1 – 8.5) to better quantify the written statements.

11. On page 6 line 33 of S.M. “ sharp decline from 2017 to 2020” I would suggest to report the value of the sharp decline
    Please refer to the response to point 10.

12. On page 7 of S.M. “small rise”.. “dropping sharply” I suggest corroborating these descriptions with quantitative values.
    Please refer to the response to point 10.