Response to Andreas Engel

We thank the reviewer for the careful review of the manuscript and study and for their detailed comments. Particularly relevant in our revised document is improvements and additions to figures in the manuscript, which address a number of comments by reviewer #3. We have also added clarifying language in places and added additional detail in the manuscript where requested. Overall, after addressing critiques and questions posed by the reviewer in the revised manuscript, we feel the manuscript has improved and the study is strengthened.

Here we detail changes made in our revised manuscript in order to address particular points for reviewer #3.

**Figure 1 isn't very nice and needs to be improved.**

We do agree that the resolution in the schematic of our GC-ECD system was poor and have updated the figure output file to improve its resolution.
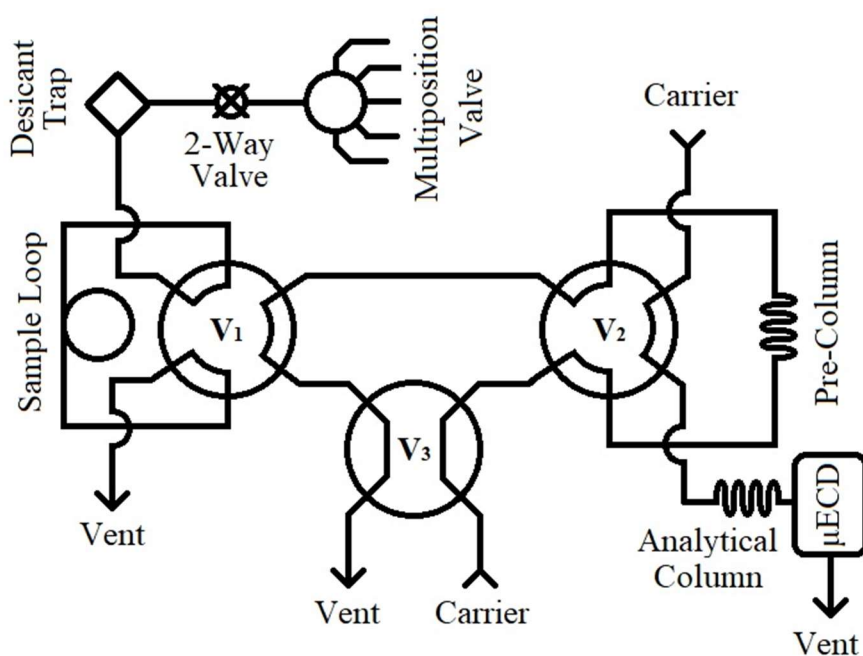


**Replace Non-Linear plots with Difference plots.**

We agree that difference plots can provide additional insight where non-linear effects are large. However, after consideration, we do feel that the measured v. expected plots shown in Fig. 4 are straightforward to interpret. For this reason, we have kept Fig. 4 as is and added additional difference plots for $N_2O$ and $SF_6$ in the supplemental documentation over the entire range measured. We also refer to these plots within the text when discussing non-linear effects.
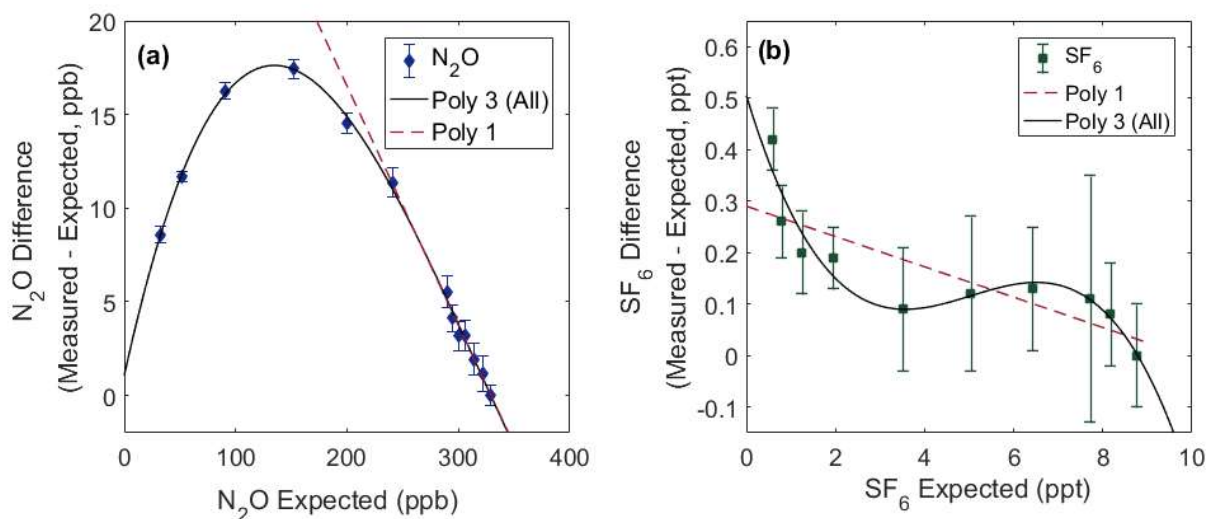
**Figure S1.** Mole fraction difference from expected plots for $N_2O$ (a) and $SF_6$ (b) detector response calibration measurements. Solid black lines are $3^{rd}$-degree polynomials fit to the whole data range. For $N_2O$, $1^{st}$-degree polynomial fit (red-dashed line) is only fit to data with mole fractions expected to be greater than 295 ppb. For $SF_6$, $1^{st}$-degree polynomial fit spans the entire data range.

**Statement on data and archive availability for other studies and the amount of air stored in the tanks.**

A statement on data availability is included at the end of the manuscript that we make all $N_2O$ and $SF_6$ data available to the scientific community upon publication. The text in section 1 (page 4, line 13) states the current air pressure in archive canisters which ranges from 60-2000 kPa bar.

**p. 1. l. 17.: is this precision for $SF_6$ not dependent on the mixing ratio, which has changed significantly during this time period?**

The precision of measurement provided in the abstract and later in the section 2 (methods) is for current ambient mixing ratios and based on repeated analyses of the NOAA reference cylinder (328.71 ± 0.5 ppb $N_2O$, 8.76 ± 0.06 ppt $SF_6$). We have modified the language in the abstract to clarify this point as suggested. For archive samples, the absolute measurement precision was determined to be relatively consistent across the concentration range measured for $N_2O$ and $SF_6$ based on replicate analysis (see error bars Fig. 5). However, relative precision of measurement for samples is dependent on the mixing ratio measured for $SF_6$ because of the wide range of mixing ratios in the archive. As noted, the result of this is the largest relative uncertainty is associated with the oldest samples where $SF_6 \leq 1$ ppt. Text in the main body now includes:

*Mean measurement uncertainty ($1\sigma$) of OHSU-PSU air archive samples for $N_2O$ is 0.23%. Mean measurement uncertainty ($1\sigma$) of $SF_6$ in the OHSU-PSU air archive samples ranges between 6.5% for samples below 1 ppt and 2.5% for samples at 4 ppt.*

**P. 1. L. 29.: please specify that the stability is only valid for these compounds. Other gases may be much more critical.**

We have updated the manuscript to include this change.

**P. 2. L. 13.: also the temporal resolution of firn samples is limited.**

We have updated the manuscript to include this change.

**P. 2. L. 17.: I think it would be worthwile to add some comments here, especially mention the best known air archive, i.e. the one from Cape Grim, incl. some references to reanalysis from air archives, e.g. from Laube, Oram and Vollmer.**

We agree with the reviewer and have updated the manuscript to include this change:

*The most well-known air archive is that of Cape Grim, Tasmania (41° S, 145° E) in the southern hemisphere, containing samples dating back to 1978 (Vollmer et al. 2018).*

**P. 2. L. 20.: a reference to the updated trend from the most recent WMO report (chapter1) could be made here).**

We agree with the reviewer and have updated the manuscript to include this change:

*The global mean mixing ratio of $N_2O$ in 2017 was 329.8 ppb with a mean annual trend of 0.85 ppb $yr^{-1}$ over the last 20 years (Dlugokencky et al. 2018).*

**P. 3. L. 20.: please include some discussion on the recent re-evalution of the $SF_6$ atmospheric lifetime e.g. by Ray et al. 2017.**

We have updated the manuscript to include this change.

**P. 5. L. 22.: I think some discussion on the reproducibility for low $SF_6$ mixing ratios is necessary here. Does it differ from those of the NOAA standards? What is the implication for the reanalysis?**

In terms of a study of the reproducibility at lower $SF_6$ mixing ratios, we were unable to perform a longer-term multi-day analyses as we did not have a sample of sufficient volume to do so (at low $SF_6$ mixing ratio). However, as the reproducibility tests at ambient $SF_6$ mixing ratios are very compatible with our analysis of the precision of measurement, we have no real reason to suspect the same study at lower mixing ratios would yield different results.

**P. 6. l1ff.: Have the authors considered a cross interference between $N_2O$ and $CO_2$? Are they separated chromatographically? If not then there could be a co-elution problem and then the dilution of the standard may result in a different matrix than in the case of air (which has shown different relative trends of $CO_2$ and $N_2O$). Co-elution of $CO_2$ and $N_2O$ may effect the sensitivity of the detector, which impacts the non-linearity correction.**

Our tests (using NDIR) indicate that $CO_2$ is well separated from $N_2O$ on a 5.5m Porapak Q column.

**P. 6. L. 23.: I think it is wrong to refer to measured $N_2O$ here; this is the "linear-response" evaluation.**

We have updated the manuscript to state "response evaluated" instead of "measured".

**P. 7. L. 3.: as above**

We have updated the manuscript to state "response evaluated" instead of "measured".

**P. 7. L. 6.: a range of values for which such a linear correction has been applied should be given here also. Please use the newest AGAGE overall reference: Prinn et al., Earth System Sci. Data, 2018**

We have updated the manuscript to include the range in values for which the linear corrections have been applied (with corrected values ranging between 298.9 – 314.8 ppb for $N_2O$ and 0.6 – 4.3 ppt for $SF_6$).

**P. 8. L. 25.: give years for the increase rates.**

Years for the rate of increase the growth rates are calculated over are provided in the first sentence of the paragraph (1978 – 1996). However, we have added "over this same time period" to the end of the sentence to improve clarity.

**P. 9. L. 13.: the ref. to Levin should be placed behind 1995.**

We have updated the manuscript to include this change.

**P. 9. L. 17.: as Edgar is largely derived from an inversion of observations, it is somewhat a circular argument to state that there is agreement.**

In the case of $SF_6$, the reviewer makes a good point here that Edgar emissions inventories are based in-part of inversion of atmospheric measurements. However, atmospheric $SF_6$ data are particularly sparse in 1980s and early 1990s, particularly in the northern hemisphere where an overwhelming majority of emissions occur. Thus, the agreement between middle latitude northern hemisphere Cape Meares, OR (45N) data and EDGAR emissions inventory is useful for updating and improving emissions inventories.

**P. 9. L. 31.: this sentence sounds funny.**

We have restructured the sentence to the following:

*Other mid-latitude northern hemisphere sites also show a seasonal phase similar to that observed at Cape Meares.*

**P.10. L. 11.: is the seasonality independent of the mixing ratios? Otherwise please give the years for which this is valid.**

So far as can be determined statistically, the seasonality observed at Cape Meares is independent of the mixing ratio.

**P. 10. L. 24.: specify what you mean by vertical diffusion here.**

We have updated the manuscript to not include vertical diffusion in the list of potential factors that may influence the observed seasonality at a location due to the ambiguity of the term.

**p.11.l. 3: check grammar on this sentence.**

We have updated the manuscript to the following:

*The analysis of archived air samples gives the mole fraction of $N_2O$ in 1980 to be $301.5 \pm 0.3$ ppb ($1\sigma$), rising to $313.5 \pm 0.3$ ppb ($1\sigma$) in 1996.*