Author Replies to Referee #1 comments on: "History of Chemically and Radiatively Important Atmospheric Gases from the Advanced Global Atmospheric Gases Experiment (AGAGE)" by Ronald G. Prinn et al.

Anonymous Referee # 1. Received and published: 1 February 2018

The referee's comments are given below in italics followed by our REPLIES and resultant paper REVISIONS in regular font. We note that this reviewer uses page and line numbers from the "FINAL SUBMIT" pdf version which are slightly different than those in the "Manuscript under review" version on the ESSDD website.

This paper reviews the past and present measurement and modelling activities of the AGAGE network, which aims to measure the concentrations and estimate the emissions of all the important species of the Montreal Protocol and non-CO2 gases of the Kyoto Protocol. The work accomplished by this group is impressive and brings a robust, reliable, and highly useful contribution to the precise and accurate monitoring of a large number of atmospheric trace gases. The paper should definitely be published after addressing the following comments and questions.
REPLY: We thank the referee for taking the time to provide a comprehensive review.

Referee General Comments:
1/The AGAGE group made the choice to report both measurement and modelling activeities. The measurement part is very convincing and related to a widely used database available on the WEB. It relies to precise and accurate observations of a large number of trace gases, useful for various scientific communities. I am less convinced by the modelling part (3.4 to 3.7) because:
(a)Interpreting observations into emissions is related to different larger uncertainties than the measurements (transport, set up of inversions, chemistry, ..), which implies to relate to many approaches to fully address uncertainties? The authors have developed several of them but it should be mentioned in the paper that other approaches exist (ex : variational approaches not mentioned, OH results from CTMs & CCMs not mentioned, . . .) which may give different results (see also some specific comments)
REPLY: Source (emission) and sink estimations are a major product of the AGAGE program, and measurement and modelling have been the two interactive pillars of the program from its inception. We agree that there are other estimation methods besides ours, but regarding OH, we note that section 4.6 in the paper does in fact refer to both the AGAGE and non-AGAGE estimates of OH levels, so they are indeed included. Also, section 4.8 includes a number of papers that use AGAGE data but not AGAGE models/methods.
REVISIONS: To make this clearer, we have changed the title of 4.8 to include "Alternative Models", and we have added the following text at the end of the opening paragraph of section 3.3: "Here, and in sections 3.4-3.7, we summarize the methods and models actually used by AGAGE scientists to interpret AGAGE measurements. There are alternative methods and models that may give differences in estimated emissions especially at regional scales. The AGAGE publications generally address the issue of differences, if any, between the estimated emissions and those reported in prior studies by other non-AGAGE scientists. Some of these alternative methods are addressed in sections

4.6 and 4.8, but it is beyond the scope of this paper to review all the alternatives. Instead we refer the reader to 2 comprehensive books that provide in-depth summaries of most of the major models and methods used to estimate sources and sinks from measurements (Enting, 2002; Kasibhatla et al, 2000)".

(b) There is apparently no database associated to the model results reported here. Then is it in the editorial policy of ESSD to report them?

REPLY: We considered providing Tables of estimated emissions on the AGAGE website, but because (as the referee also notes) the estimates depend upon the measurements, models and methods used, we did not want people using these emissions without being aware of this. We concluded that we should instead refer the reader to the many peer-reviewed publications that report the models and methods along with the estimates and make these publications available for downloading (usually as pdfs) on the AGAGE website (see section 4.9 for details about accessing the "archive" of these publications). These publications also address the issue of differences, if any, between the estimated emissions and those reported in prior studies by other non-AGAGE scientists.

(c) If maintained, these sections may be a bit more synthetized as it is looks like catalogues (maybe through tables of references per gas)?

REPLY: We considered adding a Table of references but found that it would need so many footnotes addressing the model, method and data used to make it more cumbersome to read than the text we provide. We note that sections 3.3-3.7 in total comprise less than 8% of the length of this paper, so they are already highly abbreviated.

2/The number of self-citations is high in the paper which is partly normal in such a review paper but, on the top of the remark on existing alternative modelling methods, it would be more generally fair to quote major other results, especially when they differ from AGAGE ones.

REPLY: We agree. Please see our REPLY and REVISONS regarding the related referee comment 1(a) above; we have now explicitly signaled in section 3.3, the alternatives already cited in sections 4.6 and 4.8 and listed in the References, and we now also cite 2 books covering alternative inverse methods.

3/In the measurement sections, calibration and comparison questions are a bit too spread between sections. I suggest to make these important questions about scales and comparisons more structured and clear (see also specific questions). Would it be more robust to move in the future to only one primary scale per gas whenever it is possible (e.g. main GHG gases)?

REPLY: We do not understand why calibrations (section 2.6) and inter-comparisons (section 3.2) should not be separate from one another. Calibration scales ought to be independent and de novo, not influenced by other calibration scales to which they may be compared. Perhaps this reviewer's perception is that the calibration process itself is somehow dependent on comparisons with other calibrations, but this is emphatically not true. We think it is entirely appropriate to separate these two sections. Also, multiple independent calibration scales, such as those made by AGAGE, NOAA, Tohoku, Empa, NPL, NIST and others, add strength and resilience to the research community's understanding of the accuracy of atmospheric abundances and trends. There is no single "correct" scale, for gases measured in AGAGE, just traceability and comparability. We

emphasize in section 3.2 that AGAGE data can also be reported on any other scale with which AGAGE measurements have been compared, and the resultant differences in emission and sink estimations can then be shown by repeating the inversions using the alternative scales.
REVISIONS: We add at the end of section 2.6: "Although the adoption of a single primary calibration scale from a central calibration facility for each measured species has been advocated by some researchers, AGAGE does not favor this approach. The existence of more than one independent high-precision traceable calibration scale for each measured species, with frequent inter-comparisons among independently calibrated field measurements (see Table 5, section 3.2), as well as with direct inter-comparison of the calibration standards themselves (Hall et al. 2014), reduces vulnerability to systematic error and long-term calibration drift for all participating primary calibration and measurement programs."

4/ In the measurement sections, no mention of links/comparisons is presented with the European ICOS network. This should be added, at least as perspectives.
REPLY: AGAGE data is already freely available to ICOS researchers through the AGAGE websites. Formal relations between European AGAGE stations and ICOS are up to the leaders of these stations, and so far, 3 of the 4 AGAGE European stations (Monte Cimone, Jungfraujoch, Ny Ålesund) are in the process of formally joining ICOS.
REVISIONS: We have added the following text on ICOS to the discussion of the Global Observing System in section 1.3: "AGAGE European stations provide data to, and Monte Cimone, Jungfraujoch and Ny Ålesund are now formally joining, the Integrated Carbon Observation System (ICOS) that coordinates pan-European observations of GHGs."

5/As the paper is quite dense to read, it would be good to have a short systematic description of each sub sections X.Y at the beginning of each section X. Also, in some places with lists of papers in the text, adding tables might lighten the reading.
REPLY: We agree. We already have a description of the structure of the paper in the opening of section 1 (Introduction). While this describes the 3 subsections for section 1, it provides only the overall themes for sections 2-5. Following the reviewer's suggestion, we therefore add a description of the relevant subsections in the opening paragraphs of sections 2-4 (section 5 is short with no subsections).
REVISIONS: Here are the descriptions that we now include in the opening paragraph of each section:
(Section 2) "In this section, the first 4 subsections discuss the AGAGE GC-MD (2.1), Medusa-GC-MS (2.2), optical spectroscopy (2.3), and isotopic (2.4) instruments. Then we address data acquisition and processing (2.5), instrumental calibration (2.6), primary and affiliate station facilities and infrastructure (2.7), secondary stations (2.8), and stored air archives (2.9)."
(Section 3) "In this section, the 7 subsections address: meteorological interpretation of data (3.1), data inter-comparisons (3.2), flux estimation using data and models (3.3), and then flux estimation using 3D Eulerian models (3.4), 3D Lagrangian models (3.5), merged 3D Eulerian and Lagrangian models (3.6), and simplified (2D) models (3.7)."
(Section 4) "In this section, the 9 subsections discuss: trends in Montreal Protocol gases and their replacements (4.1), whether the Montreal Protocol is working (4.2), trends in

Kyoto Protocol gases (4.3), recent rise of powerful synthetic greenhouse gases (4.4), trends in radiative forcing (4.5), determination of OH concentrations using models and multiple gases (4.6), AGAGE emission estimates for all gases (4.7), emission estimates from multiple networks, measurement platforms and alternative models (4.8), and access to AGAGE publications (4.9)."

Referee Specific comments:

P2-L19 and P5-l10: "real time" does not seem suitable for AGAGE observations. Do you mean continuous? Real time means that the observations are available for the external world at the moment they are performed. Please rephrase.

REPLY: The term "real time" does not necessarily mean "available for the external world". Through internet connections, the data is available to the AGAGE scientists in real time. Obviously, we do not release data to the public until it is validated.

REVISIONS: To avoid misinterpretation, we define what we mean by real time by changing the text at the first time this term is used on pg. 2 to: "The case for high frequency measurement networks with data available to operators in real time"…

P2-L21: "AGAGE is characterized by its capability to measure globally, at high frequency, the trends and emissions of all of the important non-carbon dioxide (CO_2) species . . .": emissions are not measured by AGAGE but estimated from AGAGE atmospheric observations. This sentence should be reformulated in this sense.

REPLY: We agree.

REVISIONS: We have added "and estimate from these trends, the" before "emissions".

P3-L10: "Affiliate stations use similar but not identical cryogenic pre-concentration GC-MS systems ": what are the main impacts of such differences?

REPLY: The main impacts are that there are some species measured by Medusas but not by Affiliates and there are small differences in precisions. These precisions are reported with the data from each instrument. As already stated, all instruments are placed on the same trace gas calibration scales.

P3-l22:" typical measurement precisions: what about the accuracy of the measurements? I guess this is very small but it would be worth reporting the magnitude of systematic errors if evaluated.

REPLY: A sentence is added on accuracy that refers the reader to section 2.6.

REVISIONS: Text added: "The accuracy of the measurements is determined by calibration scale and tertiary tank accuracies that are discussed in section 2.6."

P3-l20-27: Are there overlapping between GC and new laser spectrometers? This would of course be perfect to insure the robustness of the transition at the different sites. Please mention it if so.

REPLY: Yes. Our protocol calls for at least several months of overlap before one instrument replaces another.

REVISIONS: The following text has been added to this effect: "The GC-MD and optical spectroscopy instruments will follow the AGAGE protocol used for all cases where a new improved instrument replaces an earlier one; namely, the 2 instruments are run

together for at least several months (and e.g. years for gases currently measured on both the GC-MD and Medusa GC-MS) to ensure data comparability and verify improvements."

Table 1: Can you comment shortly in the text on the range of precisions form <1% up to 10% for some compounds?
REPLY: Good point.
REVISIONS: We add the following text where Table 1 is first mentioned: "In general the precisions in Table 1 are highest (<0.1%) for the species with highest absolute mole fractions and lowest (~10%) for those with the lowest mole fractions; there are also more subtle differences depending on the species behavior in the trapping (Medusa), separation (GC), and detection (MS, MD (ECD, FID, MRD)) stages."

P5-l12: how do you attribute local pollution events (red dots on figure 2)? From Back trajectories? Wind direction & speed at the station? maybe refer to section 3.1
REPLY: Correct. The method is described in section 3.1.
REVISIONS: We add the following sentence between "local pollution." and "Note also": "Our approach for identifying these pollution events is discussed in section 3.1."

P6-l11-13: The European ICOS is not mentioned here. What are the relation between ICOS and AGAGE?
REPLY: ICOS is now discussed in section 1.3. See our REPLY and REVISIONS above to Referee General Comment 4/.

P10-l2:"intercalibration factors": it would be good to report these factors between NOAA/AGAGE-primary/Agage-affiliated in a table. How do they vary with time?
REPLY: We agree. We now provide a Table (new Table 5) of these factors in section 3.2 (Inter-comparisons), along with text regarding their evolution over time as the instruments and absolute calibrations in each network are updated.
REVISIONS: In section 3.2 after the text "with GMD personnel are held from time to time.", we add the text: "Examples of the scale conversion factors determined from the comparison of AGAGE in situ data to NOAA flask results are given in Table 5. There is generally good consistency with time for these with some exceptions, most notably CCl_4. The CCl_4 comparison shows a trend with time from around 3.5-4.0% in 1995-2000 to approximately 1.5% in 2013-2017 (P. Krummel et al., manuscript in preparation). Because these factors are updated when additional inter-comparisons occur, we advise data users to consult the AGAGE website for possible updates." The following new Table 5 is also added to section 3.2:

Table 5: Scale conversion factors between NOAA and AGAGE (SIO) expressed as NOAA/AGAGE ratio, based on comparison of NOAA/ESRL/GMD flask data to AGAGE *in situ* data at common sites. For CH_4, N_2O and SF_6, NOAA flask data from the Carbon Cycle and Greenhouse Gases (CCGG) group have been used, for all other species NOAA flask data from the Halocarbons and other Atmospheric Traces Species (HATS) group are used. The respective scales used in each network are indicated in the table along with the instrumental method used for the analysis. The sites used in the comparisons are listed in column five, followed by the length of the comparison period. Lastly, comments on the consistency of the comparisons for each species are given.

Species	Ratio (NOAA/AGAGE)	NOAA Scale Method	AGAGE (SIO) Scale Method	Sites	Time Period	Comment
CH_4	1.0001±0.0007	NOAA-2004A GC-FID	Tohoku University GC-FID (GC-MD)	5 sites (CGO,SMO,RPB,THD,MHD)	1993-2017	0.1% consistency over time.
N_2O	0.9983±0.0005	NOAA-2006A GC-ECD	SIO-16 GC-ECD (GC-MD)	5 sites (CGO,SMO,RPB,THD,MHD)	1997-2017	0.1-0.2% consistency over time, slight increasing trend of 0.08% per decade.
SF_6	1.0049±0.0029	NOAA-2014 GC-ECD	SIO-05 GC-MS-Medusa	6 sites (CGO,SMO,RPB,THD,MHD,ZEP)	2004-2017	Small step in 2010, 0.5% consistency over time.
CFC-11	0.9993±0.0009	NOAA-2016 GC-ECD	SIO-05 GC-ECD (GC-MD)	4 sites (CGO,SMO,THD,MHD)	1993-2017	~1% consistency over time
CFC-12	0.9962±0.0010	NOAA-2008 GC-ECD	SIO-05 GC-ECD (GC-MD)	4 sites (CGO,SMO,THD,MHD)	1993-2017	0.5% consistency over time
CFC-113	1.0003±0.0023	NOAA-2003MS GC-MS	SIO-05 GC-ECD/GC-MS-Med	4 sites (CGO,SMO,THD,MHD)	1993-2017	~1% consistency over time
CCl_4	1.015-1.038 (not constant, see comments)	NOAA-2008 GC-ECD	SIO-05 GC-ECD (GC-MD)	4 sites (CGO,SMO,THD,MHD)	1995-2017	Trend: 3.5-4.0% difference in 1995-2000, to approximately 1.5% difference in 2013-2017.
CH_3CCl_3	1.0055±0.0109	NOAA-2003 GC-MS	SIO-05 GC-ECD/GC-MS-Med	4 sites (CGO,SMO,THD,MHD)	1993-2017	Initial trend during 1993-2000, from 3% down to 0.5% difference, then good agreement within 1%.
HCFC-22	0.9971±0.0027	NOAA-2006 GC-MS	SIO-05 GC-MS-ADSMed	4 sites (CGO,SMO,THD,MHD)	1998-2017	1-2% consistency over time
HCFC-141b	0.9941±0.0049	NOAA-1994 GC-MS	SIO-05 GC-MS-ADSMed	4 sites (CGO, SMO, THD, MHD)	1998-2017	~2% consistency over time
HCFC-142b	0.9743±0.0052	NOAA-1994 GC-MS	SIO-05 GC-MS-ADSMed	4 sites (CGO, SMO, THD, MHD)	1998-2017	~2% consistency over time
HFC-134a	1.0015±0.0048	NOAA-1995 GC-MS	SIO-05 GC-MS-ADSMed	4 sites (CGO, SMO, THD, MHD)	1998-2017	~2% consistency, better recently
HFC-152a	0.9976±0.0227	NOAA-2004 GC-MS	SIO-05 GC-MS-ADSMed	4 sites (CGO, SMO, THD, MHD)	1998-2017	2-3% consistency over time
H-1211	0.9799±0.0050	NOAA-2006 GC-MS	SIO-05 GC-MS-ADSMed	4 sites (CGO, SMO, THD, MHD)	1998-2017	~2% consistency over time
H-1301	0.9766±0.0098	NOAA-2006 GC-MS	SIO-05 GC-MS-Medusa	3 sites (CGO, SMO, THD)	2004-2015	~2% consistency over time
H-2402	1.0208±0.0100	NOAA-1992 GC-MS	SIO-14 GC-MS-Medusa	4 sites (CGO, SMO, THD, MHD)	2004-2017	Small step change 2008-2009, 3-4% consistency over time
CH_3Cl	1.0074±0.0073	NOAA-2003 GC-MS	SIO-05 GC-MS-ADSMed	4 sites (CGO, SMO, THD, MHD)	1998-2017	2% consistency over time

Table notes:
Comparisons between NOAA HATS data and AGAGE in situ were performed based on the NOAA data posted on the ftp site: ftp://ftp.cmdl.noaa.gov/hats/
GC-MS-ADSMed indicates data from the ADS instruments at Cape Grim and Mace Head used from 1998-2003, with Medusa data used from 2004 onwards at the sites indicated.
GC-ECD/GC-MS-Med indicates a combined data record from the GC-ECD (GC-MD) instruments with the GC-MS-Medusa data used for the latter part of the record.
Sites: CGO – Cape Grim, Australia; SMO – Cape Matatula, Samoa; RPB – Ragged Point, Barbados; THD – Trinidad Head, USA; MHD – Mace Head, Ireland; ZEP – Zeppelin Mountain, Ny-Ålesund, Norway.
Some species are measured by multiple instruments and/or flask samples, selected results shown here.

P10-l6-7: Do H_2O corrections are really efficient compared to drying? More should be said on this issue. The choice made in practice should be indicated in table 4.
REPLY: As it says in the text below Table 4, our experience is that drying the air sample decreases the needed H_2O interference correction and, not surprisingly, increases the precision of the data. The instruments and stations in Table 4 with "E.N. Driers" have adopted this technology.
REVISIONS: Text has been added to the Table 4 caption to make this clear: "Instruments with Earth Networks (EN) driers lower the sample water vapor mole fractions to decrease H_2O interference."

P11-l21: "a precision of at least 0.3 per mil (‰)" for individual measurement? or after averaging? How long?
REPLY: Precision of at least 0.3 per mil is for individual measurements spanning 28 min.
REVISIONS: Text added on this: "for individual measurements spanning 28 min. For at least 0.1 per mil (‰), we need to average 3-11 such measurements depending on the isotope (Harris et al, 2013)."

P12: do the GCWerks package keep track of raw data can eventually correct the full time series back in time if necessary?
REPLY: Yes it does.
REVISIONS: The following text is added on this at the end of paragraph 3 in section 2.5: "GCWerks also keeps all of the raw data, including the chromatograms, thus enabling routine reprocessing of the entire record for each species at each station whenever needed (e.g. when calibration scales are updated (see section 2.6) or when peak integration methods are improved)."

P13: The question of comparison of scales and eventual drifts and corrections between networks is an issue for me and deserves more attention. A table? Do comparisons between networks change with time? also treated in 3.2. It might be more clear to regroup this part with 3.2 or at least refer here to 3.2.
REPLY*:* See our REPLY and REVISIONS to P10-l2 above. A Table 5 and text has been added to section 3.2.
REVISIONS: We add the following text here: "This subject of inter-calibration is discussed further in section 3.2".

P14-section 2.9: do these canisters drift with time for some species? Has it been investigated?
REPLY: Drifts are monitored in all stored tanks (calibration, air archives, etc.) usually by inter-tank comparisons (see the discussion about drifts in section 2.6 regarding the R1 scales). Only rarely do drifts occur for a few tanks and a few of the more chemically active species. These details are discussed when necessary in the papers using the real time and archive air samples (see sections 2.6 and 2.9).

P15-l1-2: please provide a more detailed description of the algorithm (1-2 sentences)? Does it do a frequency analysis? change magnitudes?
REPLY: There are already several sentences and 4 references for our statistical pollution identification algorithm in section 3.1. We therefore presume that the Referee is referring to the way the NAME model is used to test the statistical algorithm.
REVISIONS: We replace the brief text between "to further evaluate" and "NAME back trajectories" with the following expanded text: "the statistical pollution algorithm (O'Doherty et al., 2001, Cunnold et al, 2002) and include it as part of the pollution/background identification flag associated with each measurement. The NAME model is Lagrangian (section 3.5), where large numbers of particles at the station are effectively advected backwards in time by 3D reanalysis meteorological fields, with turbulent dispersion represented by a random walk technique. Particles first encountering the surface or surface boundary layer in known trace gas emitting regions are then

flagged as polluted. An observation is also considered potentially polluted if the atmosphere at the station is stable with very low winds and known nearby trace gas sources."

Section 3.2: No mention is made of the European ICOS network. Please provide at least a strategy of comparison if none is done at the moment as ICOS will structure EU GHG observations in the future.
REPLY: See our REPLY and REVISIONS to the related General Comment 4/. While we are not in a position to advise ICOS, it would of course be advantageous if ICOS were to fund inter-comparisons between European AGAGE stations (on AGAGE calibration scales) and the other stations in ICOS (on their scales). That way, the multi-station ICOS datasets can be placed on any of the scales involved. On this latter topic, see our reply to General Comment 3/.

P15-l47-48: a box model is not a CTM for me as transport is treated crudely, but more a conceptual model. Please rephrase.
REPLY: Well, this is a matter of semantics. For us, our 12-box model is a 2D CTM (chemical transport model) that includes advective and eddy diffusive transport with the transport parameter patterns taken from observed zonal-average wind and wind variance data and parameter values optimally estimated to fit observed near-inert trace-gas data on horizontal (north-south) gradients in the troposphere and vertical (troposphere-stratosphere) gradients. We do refer to this model in section 3.7 as a "simplified model" so we are not over-stating its attributes. We have also called it a simplified 2D CTM in prior peer-reviewed papers.

P16: H is assumed linear as you write it, although many of the species measured are chemically active, potentially introducing non linearities, as mentioned just after. Please reformulate the first paragraph of page 16 to make this more clear start with general formula with H(x), introduce the linear hypothesis and then the weal-linearity sentence.
REPLY: We agree and have added text appropriately.
REVISIONS: The following is added as the 4th sentence of the second paragraph of 3.3: "Formally, since the chemical lifetime for a reactive trace gas can depend on emissions of that gas, then $\mathbf{H = H(x)}$ so the equation $\mathbf{y = Hx}$ is nonlinear in $\mathbf{x}$. For many ozone-depleting and greenhouse gases this nonlinearity is negligibly weak and is ignored. Exceptions exist as discussed later."

P16: what about the covariances in the R matrix ?
REPLY: See our REPLY and REVISIONS to the very similar comment *P16* below.

P16-l4-5? Isn't P should be Pprior in this formula?
REPLY: Yes.
REVISIONS*:* Change made.

P16-l5-9: When moving from AGAGE observations to the use of AGAGE observations in modelling, the authors should not limit to self-citations or only citations from close collaborators of AGAGE, but open a bit citations as several groups around the world

use atmospheric data (including AGAGE) to estimate trace gas emissions (especially for GHGs). Examples : Houweling or Saunois papers for methane, Montzka or Krol papers for CH3CCl3, . . .
REPLY: The Montzka and Krol papers for CH_3CCl_3 were already cited in section 4.6. The two Saunois et al (2016, 2017) papers were already in the Reference list and were supposed to have been cited in section 4.8. They are cited now. Please also see our REPLY and REVISIONS regarding the related referee comment 1(a) above; we have now explicitly signaled in section 3.3, the non-AGAGE-led papers already cited in sections 4.6 and 4.8 and listed in the References.
REVISIONS: The following texts are added in section 4.8 as the second sentence and the second-to-last sentence respectively: "A number of the multi-network studies also applied alternative models and inverse methods to those used in AGAGE (sections 3.4-3.7)"……."Saunoir et al (2016, 2017) used multi-network data and alternative models to elucidate the 2000-2012 methane budget and its multi-year variability."

P16-l30: for the aggregation error please quote Kaminski et al., 2001.
REPLY: Yes. Thank you for the reminder.
REVISIONS: Citation added here, and paper placed in References section:
"Kaminski, T., P.J. Rayner, M. Heimann, and I.G. Enting. On aggregation errors in atmospheric transport inversions. *J. Geophys. Res.*, 106, 4703-4715, 2001."

P16: did you address of diagonal terms of P and R (error covariances?)
REPLY: Yes. We add some brief text on this at the end of section 3.3.
REVISIONS: We add: "Non-zero values for the off-diagonal elements of **R** and **P** can occur. Because the AGAGE measurement stations are well separated, off-diagonal elements (co-variances) of **R** should be much smaller than the diagonal elements (variances) and are usually ignored. Also, **P** element co-variances should be much smaller than the variances except when state vector elements are correlated which can be avoided when choosing the elements. These co-variances (off-diagonal elements of **P**) are discussed in the individual papers where they are relevant."

P17, section 3.5: Why moving from Hysplit to LPDMs? Please provide minimum differences/improvements.
REPLY: We did not intend to say that one model is better than another. They all have their strengths.
REVISION: On line 7 of 3.5 we replace "but now utilize" with "and now we also use additional"

P17-l43: "unless an adjoint model of the CT is available": please provide a reference, e.g. Chevallier et al., 2005 or Meirink et al., 2008.
REPLY: Yes. Thank you.
REVISIONS: We now cite here (and add to the Reference section) the later paper:
"Meirink, J. F., P. Bergamaschi, and M. C. Krol, Four dimensional variational data assimilation for inverse modelling of atmospheric methane emissions: method and comparison with synthesis inversion. *Atmos. Chem. Phys*., 8, 6341-6353, 2008, atmos-chem-phys.net/8/6341/2008/."

P18-l18: "is well suited to full uncertainty": I would replace full by extensive as one can hardly address transport model errors with a box model but allows to do a lot of simulations because of its low computing cost. Please rephrase.
REPLY: In response to a comment from Referee #2, we have replaced the second sentence in section 3.7 with: "Therefore, 2D models have been widely used to analyse long-term trends in AGAGE data. The AGAGE 12-box model (Cunnold et al.,1994; Prinn et al., 2001, 2005; Rigby et al., 2013, 2014) uses transport parameters that have been "tuned" using AGAGE observations of trends and latitudinal gradients (e.g. Cunnold et al., 1994; Rigby et al., 2013) so that the model can simulate monthly mean observations at background AGAGE stations with pollution events removed. From these simulations, multi-decadal AGAGE time series have been used to estimate trace gas global emissions and atmospheric lifetimes."
We agree that "full" is inappropriate. We disagree that uncertainty in transport is not addressed with this 2D model. The advective and eddy-diffusive terms and their uncertainties in the 12-box model are optimally estimated to fit inert trace gas distributions, so transport errors can be included in Monte Carlo studies (e.g. Huang et al, 2008).

P19-l10: "although CFC-11 emissions post-2010 are rising, Figure 4": how did these emissions have been inferred? Which inversion method, reference paper? Do other inversions infer similar results?
REPLY: We use the standard 12-box model and inversion method (multiple references given in section 3.3 and 3.7). The results are robust in that the global average AGAGE measurements clearly show a slowing of the rate of decrease of CFC-11 in recent years.
REVISIONS: To clarify, we add on line 10 after the word "Figure 4" the text: ", the method used does not provide regional-level emission estimates needed to identify causes of this rise. Montzka et al (2018) recently concluded that East Asia was the source.", we add to the Fig. 4 caption after the word "emissions" the text: "(using our Bayesian statistical approach (section 3.3) and our 12-box model (section 3.7)).", and finally we add to the Reference section:
"Montzka, S.A., G.S. Dutton, P. Yu, E. Ray, R.W. Portmann, J.S. Daniel, L. Kuijpers, B.D. Hall, D. Mondeel, C. Siso, D.J. Nance, M. Rigby, A.J. Manning, L. Hu, F. Moore, B.R. Miller, J.W. Elkins, A persistent and unexpected increase in global emissions of ozone-depleting CFC-11, *Nature*, in press, 2018."

Section 4.2: I suggest to change the order of the points to put first direct results from AGAGE observations (2, 4, 5, 6) and then the analyses (1, 3?).
REPLY: While we agree that there are alternative reasons to use the suggested order, given the title of section 4.2 (Is the Montreal Protocol working?) we prefer to put (1) and (2) first.

P20-l14: GHG! anthopogenic GHG.
REPLY: We agree.
REVISIONS: "anthropogenic" added

P20-l21: idem for N2O
REPLY: Not sure what is being requested here. Chemical formula for nitrous oxide is defined several times beginning in the Abstract.

P21-l4: how many lead authors & contributing authors? may be worth précising this.
REPLY: We think there is too much detail needed to provide these numbers in a meaningful way. For example, AGAGE scientists, AGAGE data and AGAGE modeling results played a prominent role in the WMO-UNEP 2010 and WMO-UNEP 2014 Ozone Assessments, providing coordinating Lead authors (1in 2010, 1in 2014), Co-authors (2 in 2010), contributing authors (7 in 2010, 6 in 2014) and reviewers (8 in 2010, 6 in 2014). AGAGE measurements, model results and scientists also played a prominent role in the 2013 SPARC Report No. 6 of the WCRP on the lifetimes of ODSs, their replacements, and related species providing an Editor, a Lead Author, 3 Co-Authors, and 2 Reviewers. The AGAGE-led paper on the re-evaluation of the lifetimes of the major CFCs and CH_3CCl_3 using atmospheric trends (Rigby *et al.*, 2013), was an important input into both the 2013 SPARC Report and the 2014 Ozone Assessment. AGAGE played similar significant roles in the IPCC 4th and 5th Assessments: Climate Change 2007 and 2013, providing data, modeling results, Lead authors (1in 4th), Contributing authors (3 in 4th), and Reviewers for WG1, Chapter 2 (4th & 5th), and WG1, Chapter 6 (5th). And so on.

P21-l18: what GWP has been used to get CO2-equivalent emissions? is it for a 100-year horizon? Please precise.
REPLY: Our omission.
REVISIONS: This text is now added after "CO_2-equivalent emissions": "using 100-year GWPs".

P23-l10-11: for HCHC22 Fortems-Cheiney et al. (2013) can be quoted, as can be Fortems-Cheiney et al. (2015) for HFC-134a
REPLY: We agree.
REVISIONS: These citations now added here, and the 2013 paper added to the Reference section (2015 paper already there): " Fortems-Cheiney, A., F. Chevallier, M. Saunois, I. Pison, P. Bousquet, C. Cressot, R. H. J. Wang, Y. Yokouchi, and F. Artuso. HCFC-22 emissions at global and regional scales between 1995 and 2010: Trends and variability. *J. Geophys. Res.: Atmosp*heres, 118, no. 13, 7379-7388, 2013."

P23: it should be quoted here that MCF-based proxy methods to retrieve OH generally infer larger IAV than CTM or CCM calculations (e.g. Montzka et al., 2011; Voulgarakis et al., 2013; Naik et al., 2013)
REPLY: We agree.
REVISIONS: We add in the first paragraph of section 4.6: "Generally, inter-annual variability in OH inferred from CH_3CCl_3 inversions are larger than those calculated in atmospheric photochemical models (e.g. Montzka et al, 2011), and the reasons are currently unresolved."

P24-l9, . . . and tranport model errors
REPLY: Text added.
REVISIONS: We add after "and" on line 9: "in chemical transport models including"

P24-l15: "to help resolve this issue" may be replace by: "to quantify such unreported emissions"
REPLY: Agreed.
REVISIONS: Text changed on line 15 to: "to better quantify these unreported emissions."

P24-l23: "Recent": ambiguous here as 2008 was ten years ago. Please be more precise.
REPLY: Agreed
REVISIONS: Changed "recent" to "2007"

Author Replies to Referee #2 comments on: "History of Chemically and Radiatively Important Atmospheric Gases from the Advanced Global Atmospheric Gases Experiment (AGAGE)" by Ronald G. Prinn et al.
Anonymous Referee # 2. Received and published: 6 March 2018

The referee's comments are given below in italics followed by our REPLIES and resultant paper REVISIONS in regular font. We note that this reviewer uses page and line numbers from the "Manuscript under review" version on the ESSDD website which are slightly different from those in the "FINAL SUBMIT" pdf version.

General Comments:
This is an overall well written and informative overview of the status of the AGAGE project. It is a timely publication, given that it has been some 18 years since the last overview published by Prinn et al. This is a very impressive project, led by the world leading experts in this field. Measurements and data are of the highest quality that are achievable with current technologies. Data are disseminated to the global community and used around the globe. There is a bit of redundancy in places, with the same/similar material/text being repeated in different sections.
I recommend publication of this manuscript after reasonable consideration of the comments listed below that may yield some further improvements to the overall nicely written document.
REPLY: We thank the referee for taking the time to provide a comprehensive review.

Please comment on the connection/relationship between AGAGE and the World Meteorological Organization Global Atmospheric Watch programme.
REPLY: AGAGE is an active contributor to GAW and, as discussed in section 5, AGAGE data are provided to the GAW World Data Center for Greenhouse Gases (WDCGG) website. Discussions about an even more formal relation between AGAGE and GAW are ongoing. We have now included WMO-GAW in the discussion of the Global Observing System in section 1.3.
REVISIONS: We add in section 1.3: "AGAGE contributes to the World Meteorological Organization's Global Atmosphere Watch (WMO-GAW) and regularly provides its data to the WMO-GAW's World Data Center for Greenhouse Gases (WDCGG) website (see section 5)."

Specific Comments:
Page 2/Line 46: .....accomplishments, and ....
REPLY: Thanks.
REVISIONS: Comma added.

Figure 1: It would be nice to get a clearer description/definition of what criteria are applied for stations to be included as 'AGAGE Station' versus 'AGAGE Affiliate Station'. And what about other sites/programs in the world where related monitoring occurs? For instance, I saw at a recent visit to the Zugspitze Station that the German Umweltbundesamt Agency is conducting CFC monitoring at that site. What other

programs are there that capture and report these same or a subset of these compounds? It would be nice to at least include a more inclusive summary/comparison of the global monitoring activities of these gases outside of AGAGE.
REPLY: As already stated in sections 1.1, 1.2 and 2.2, the AGAGE stations (primary, affiliate) are characterized by having automated GC-MS instruments with air sample pre-concentration, measuring on-site at high frequency and reporting data (directly or indirectly) on the AGAGE calibration scales. Some stations also have additional instruments (see sections 2.1, 2.3 and 2.4). Also, the station instrumental attributes and primary and affiliate names have evolved over time. So it is difficult for us to provide further clarity. Regarding other networks, we already list in section 1.3 on the Global Observing Network both NDACC and NOAA-ESRL as related ground-based trace gas networks, and now following Referee#1's request, we have added the Integrated Carbon Observation System (ICOS) that coordinates pan-European observations of GHGs to that list. The Zugspitze station is presumably a part of ICOS.

4/Table: What does the superscript '4' in the last column heading refer to?
REPLY: Thanks for noticing this. It is a remnant from a prior version.
REVISIONS: Superscript 4 deleted.

4/Table: This table lists a number of non-methane hydrocarbons (NMHC) as well, though there is no mention/discussion in the text about NMHC being included in the AGAGE suite of measurements?
REPLY: The 4 NMHC listed in Table 4 (ethane, propane, benzene, toluene) are optional compounds measured at some of the stations. Also, CO and H_2 are measured at 2 stations. Our examples in Section 4 deliberately focus on greenhouse and ozone depleting gases, but we neglected to at least briefly discuss these other gases, and we now do so.
REVISIONS: In the opening paragraph of Section 4 (following the sentence added in response to Referee #1's General Comment 5) we add: "We focus on the greenhouse and ozone-depleting gases in this section, but note that the 4 non-methane hydrocarbons listed in Table 1 (ethane, propane, benzene, toluene) are optional compounds measured at some of the stations (for examples see Yates et al, 2010; Grant et al, 2011; Derwent et al, 2012; and Lo Vullo et al, 2015, 2016). Also, CO and H_2 are measured at 2 stations (e.g. Xiao et al, 2007). When correlated with the other gases in Table 4, these can be used as indicators of the sources of these other gases, and they are all also relevant to the fast photochemistry of OH." And we add the References:
"Lo Vullo E., Furlani F., Arduini J., Giostra U., Graziosi F., Cristofanelli P., Williams, M. L., Maione, M., 2016. Anthropogenic non-methane volatile hydrocarbons at Mt. Cimone (2165 m a.s.l, Italy): Impact of sources and transport on atmospheric composition, *Atmospheric Environment,* 140, 395–40. doi: 10.1016/j.atmosenv.2016.05.060
Lo Vullo E., Furlani F., Arduini J., Giostra U., Cristofanelli P., Williams, M. L., Maione, M., 2016. Non-Methane Volatile Organic Compounds in the Background Atmospheres of a Southern European Mountain Site (Mt. Cimone, Italy), Annual and Seasonal Variability, *Aerosol and Air Quality Research,* 16/3, 581-592. DOI: 10.4209/aaqr.2015.05.0364

Yates, E.L., R.G. Derwent, P.G. Simmonds, B.R. Greally, S. O'Doherty and D.E. Shallcross, The seasonal cycles and photochemistry of C2–C5 alkanes at Mace Head, *Atmos. Environ.*, 44, 2705-2713, doi: https://doi.org/10.1016/j.atmosenv.2010.04.043 , 2010.

Grant, A., E.L. Yates, P.G. Simmonds, R.G. Derwent, A.J. Manning, D. Young, D.E. Shallcross and S. O'Doherty, A five year record high-frequency in situ measurements of non-methane hydrocarbons at Mace Head, Ireland, *Atmos. Meas. Tech.*, 4, 955-964, doi: https://doi.org/10.5194/amt-4-955-2011, 2011.
Derwent, R.G., P.G. Simmonds, S. O'Doherty, A. Grant, D. Young, M.C. Cooke, A.J. Manning, S.R. Utembe, M.E. Jenkin and D.E. Shallcross, Seasonal cycles in short-lived hydrocarbons in baseline air masses arriving at Mace Head, Ireland, *Atmos. Environ.*, 62, 89–96, doi: https://doi.org/10.1016/j.atmosenv.2012.08.023, 2012.

4/Table: Please explain how the 'precision' that is listed here was determined. An additional column that lists the overall measurement uncertainty (resulting from precision and accuracy) would be highly desirable.
REPLY: The precisions are determined continuously from the interspersed measurements of the on-site station calibration tanks and are reported along with the mole fractions at the AGAGE data archives. The general accuracies of the AGAGE calibrations are discussed in section 2.6 and the differences between AGAGE and ESRL-GMD calibrations (section 3.2, Table 5) are an additional measure of accuracy. However, we do not combine precisions (random errors) with accuracies (systematic errors) since they are statistically distinct and need to be handled separately in estimating emissions and sinks from the measurements.
REVISIONS: We add the following text above Table 1: "The precisions for each species are determined from the interspersed measurements of the on-site station calibration tanks and are reported along with the mole fractions of the interspersed atmospheric measurements in the AGAGE data archives."

5/1: Can you be more specific than 'spectroscopic'?
REPLY: They utilize laser spectroscopy. We could provide more information here, but the later sections 2.3 and 2.4 discuss these detectors in detail.
REVISIONS: "We add "laser" before "spectroscopic".

5/36-46: It would be good to also mention the Network for the Detection of Atmospheric Composition Change (NDACC, www.ndacc.org).
REPLY: We already specifically mention NDACC on pg. 5 lines 40-41 (section 1.3).
REVISIONS: We now add: "AGAGE is an NDACC Cooperating Network" to that text.

6/Figure 2: These plots are so small that their value is rather questionable. When data labeled 'red' are for 'polluted air', shouldn't then the data for all species at a given (polluted) time be labeled consistently that way?
REPLY: This Figure addresses up front the key issue about why AGAGE demands its signature high frequency sampling to resolve these important events. Figure 1 is readable and shows that most gases measured show very frequent pollution events, and that they are often but NOT always correlated. However, the referees comment reminds us that we

should have made it clear that the definition of pollution used here is done gas by gas.
REVISIONS: We add text to the caption of Fig. 2: "Note pollution events are defined separately for each gas due to their often differing sources."

6/7: It seems a bit odd that this section starts here, after data have already been presented in Section 1/Figure 2. Maybe the best solution is to just remove Figure 1 from the introduction section, and then instead have a blown up time series graph of fewer species presented and discussed later on for demonstrating the value of high time resolution data.
REPLY: We strongly prefer to introduce the instruments, the gases measured (Table 1), and samples of the actual data obtained (Figure 2) in the Introduction (section 1.2), and not to postpone introduction of these critical subjects to later sections. See also our reply to comment 6/Figure 2.

7/33: A description of the sampling line/material/flow condition/inlet filter/line heating would be good to also include in the Instrument Section.
REPLY: These issues are addressed later in Section 2.7. Because they differ significantly from station to station (depending on the non-AGAGE measurements also made at the site, the local climate conditions, the tower setup, etc.), we have not provided details for each site in this paper. The general setup for each instrument was provided by Prinn et al (2000) and Miller et (2008), and we now refer the reader to those papers.
REVISIONS: We add after the first sentence in section 2.7: "The details about the general air sampling setup for each instrument are provided in Miller et al (2008) and Prinn et al (2000). The sampling lines are either stainless steel or layered polyethylene-aluminum-Mylar (Dekabon© or Synflex©). For more information on individual stations, we refer the reader to the AGAGE website (agage.mit.edu)."

7/35: The wording of this sentence is somewhat odd: ....increases ...being added ....
REPLY: Agreed. We rewrite this sentence.
REVISIONS: The replacement sentence is: "Instrument development work beyond Miller et al (2008) and Arnold et al (2012) continues, with enhanced operational parameters, upgrades and new species being added over time."

7/39: 'GWP' hasn't been defined. Further, I suggest to reword the whole term 'high-GWP electronics industry chemical NF3'.
REPLY: GWP is best defined later on (section 4.4) in response to a Referee #1 comment. We reword this text here as noted below.
REVISIONS: We replace "high-GWP electronics industry chemical NF_3" with "the powerful greenhouse gas NF_3 emitted by the electronics industry"

8/47: Suggest deleting the word 'single'.
REPLY: Agreed.
REVISIONS: Deleted "single".

9/15: Again, as mentioned earlier, what does it take for a site to be an 'AGAGE program site', versus being an 'Affiliate Station', and why are other sites where global CFC

monitoring is ongoing not affiliated or included?
REPLY: Agreed. See our reply on this under your *Figure 1* specific comment.
REVISIONS: See our revisions resulting from this *Figure 1* comment.

10/1-8: It may be worth mentioning also that these instruments do not require a carrier gas supply, which eases their operation.
REPLY: Agreed. But implicit in "no chromatography".
REVISIONS: Add "so no carrier gases needed" after "no chromatography"

10/14: Please explain or provide reference for 'Allan' variance analyses.
REPLY: We add references as requested.
REVISIONS: After "analyses" on line 10/15, we add: "(Allan, 1966; Werle et al., 1993)".
In the References section, we add:
"Allan, D. W., Statistics of atomic frequency standards, *Proceedings of the IEEE*, 54, 221-230, 10.1109/PROC.1966.4634,1966
Werle, P., R. Mücke and F. Slemr, The limits of signal averaging in atmospheric trace-gas monitoring by tunable diode-laser absorption spectroscopy (TDLAS), *Appl. Phys. B*, 57, 131-139, 1993"

10/Table 4: 'Station' instead of 'Stations'
REPLY: We keep "stations" since both DECC network and NPL have stations.

10/Table 4: Explain abbreviation 'EN'.
REPLY: EN refers to "Earth Networks" as mentioned in the text following the Table.
REVISIONS: Text defining EN now added to Table 4 caption in response to the related comment from Referee #1.

11/1-27: Since these measurements are not really fully integrated into the AGAGE network (yet), this section seems to be pre-mature. I suggest deleting it entirely.
REPLY: We are puzzled by this request. As noted in the text, the MIT-Aerodyne instrument has been deployed at Mace Head (temporarily back at MIT for upgrades), the Empa instrument is used for Jungfraujoch flask samples, and the CSIRO instrument is operating at Cape Grim. We think most readers will want to know about these new instruments emerging in AGAGE.

12/1: Wording seems odd: '...has continued to be developed ....'
12/10: Same here.
REPLY: We will simplify these.
REVISIONS: We change "has continued" to "continue" in both places.

12/37-41: Please provide a bit more background info why/how it matters which of these two different standard preparation methods is used.
REPLY: The method of internal addition to "zero-air" was necessary for CF_4 and NF_3 as their analytical separation is highly dependent on the presence of Krypton and other inert gases which are not present in "zero air".
REVISIONS: After the sentence ending in "agreement." on line 42, we add the sentence:

"For the volatile gases like CF_4 and NF_3, the use of the internal additions method is particularly valuable to avoid biases in their separation or detection due to interferences due to the presence of krypton and other inert gases in real air but not in artificial "zero air"."

13/42: ....at remote sites
REPLY: We reword but want to keep the adjective "more".
REVISIONS: We change "in" to "at" for both places it occurs on this line.

15/9: Would 'modelers' be a better term than 'theoreticians'?
REPLY: Some people object to being called "modelers", so we reword accordingly.
REVISIONS: We change "by theoreticians" to "in modeling".

16/0: It seems a bit odd that the statement 'briefly below' is then backed up with a series of references?
REPLY: We delete un-needed text as noted.
REVISIONS: Delete "which are described briefly below"
.
18/30-31: I think a few commas are missing before and after 'respectively'.
REPLY: We agree.
REVISIONS: Commas added after first "respectively" and after "per year".

18/34: ...have continued to rise in .....
REPLY: Agreed.
REVISIONS: Change "continue" to "have continued".

19/10: I for sure would like to know why CFC-11 emissions are rising again?
REPLY: We are currently working on this using 3D model inversions. The results in Fig. 4 were obtained using our 2D 12-box model which does not provide regional/national-level emission estimates that are needed to pinpoint specific causes. In response to referee #1on this, we added on line 10 after the word "Figure 4" the text: ", the method used does not provide regional-level emission estimates needed to identify causes of this rise. Montzka et al (2018) recently concluded that East Asia was the source.", we add to the Fig. 4 caption after the word "emissions" the text: "(using our Bayesian statistical approach (section 3.3) and our 12-box model (section 3.7)).", and finally we add to the Reference section: "Montzka, S.A., G.S. Dutton, P. Yu, E. Ray, R.W. Portmann, J.S. Daniel, L. Kuijpers, B.D. Hall, D. Mondeel, C. Siso, D.J. Nance, M. Rigby, A.J. Manning, L. Hu, F. Moore, B.R. Miller, J.W. Elkins, A persistent and unexpected increase in global emissions of ozone-depleting CFC-11, *Nature*, in press, 2018."

19/15: Are these halogenated gases included in the calculation of total stratospheric chlorine?
REPLY: Yes.

19/22: Please explain a bit more what is going on with CCl4.
REPLY: We agree. Text has been added and relevant papers cited.

REVISIONS: We add to section 4.2 (4) the text: "The 2010 and 2014 WMO Scientific Assessments of Ozone Depletion noted that emissions of CCl_4 inferred from AGAGE and NOAA observations were substantially higher (~ 50 Gg/yr) than estimates based on consumption reported to UNEP (Montzka et al, 2011a; Carpenter et al, 2014). Recent studies have attempted to re-evaluate the global CCl_4 budget (Liang et al., 2016). Estimates of the soil and ocean partial lifetimes have been revised upward (Rhew and Happell, 2016; Butler et al., 2016) and several new industrial sources have been identified (Sherry et al., 2017), substantially reducing the gap between top-down and bottom-up estimates (Chipperfield et al., 2016)."
We add to the References:
"Butler, J. H., Yvon-Lewis, S. A., Lobert, J. M., King, D. B., Montzka, S. A., Bullister, J. L., Koropalov, V., Elkins, J. W., Hall, B. D., Hu, L. and Liu, Y.: A comprehensive estimate for loss of atmospheric carbon tetrachloride (CCl_4) to the ocean, *Atmospheric Chemistry and Physics*, 16(17), 10899–10910, doi:10.5194/acp-16-10899-2016, 2016.
Rhew, R. C. and Happell, J. D.: The atmospheric partial lifetime of carbon tetrachloride with respect to the global soil sink: CCl_4 Soil Sink and Partial Lifetime, *Geophysical Research Letters*, 43(6), 2889–2895, doi:10.1002/2016GL067839, 2016.
Sherry, D., McCulloch, A., Liang, Q., Reimann, S. and Newman, P. A.: Current sources of carbon tetrachloride (CCl_4) in our atmosphere, *Environmental Research Letters*, doi:10.1088/1748-9326/aa9c87, 2017."

19/32: ....AGAGE data, and ....
REPLY: Agreed.
REVISIONS: Comma added between "data" and "and".

20/23: ....concentration has been ....
REPLY: Agreed.
REVISIONS: Change "concentrations have" to "concentration has"

21/20: Please explain what 'Annex-1' countries are.
REPLY: We provide a brief definition that suffices for this paper. There is a large literature on the requirements for the Annex-1 signatories to the UNFCCC.
REVISIONS: We add after "countries" the text "that are signatories to the United Nations Framework Convention on Climate Change (UNFCCC)".

23/10: I suggest rephrasing this sentence. Maybe: "Using HCFC-22 data for estimating the mean OH concentration yields similar results ....."
REPLY: OK.
REVISIONS: We replace "Another------similar" with "Using HCFC-22 measurements for estimating the average OH yields similar results"-----

23/21: .....significantly from year to year, but ....
REPLY: OK.
REVISIONS: Comma added between "year" and "but".

23/29: ....OH concentrations, and CH3CCl3 emissions from ……

REPLY: Thanks.
REVISIONS: We add "CH_3CCl_3" before "emissions".

24/5: ….are then critically compared …..
REPLY: Thanks.
REVISIONS: We delete "be".

24/18-19: This reads a bit odd. How can a 2005 paper be called an 'earlier study' when actually earlier data are being compared with that study?
REPLY: Agreed.
REVISIONS: We replace "earlier" with "Krol et al. (2003)"

24/23: Similar issue here. A paper published in 2008 can not really report on the 'recent' increase in methane. Maybe it reported the 'onset' of the renewed growth of methane.
REPLY: Thanks. This was also noted by Referee #1
REVISIONS: We changed "recent" to "2007".

24/28: …nitrous oxide, and Xiao …..
REPLY: OK.
REVISIONS: Comma added after "oxide".

24/30: It would be nice to learn a bit more about how the interhemispheric gradients in these beautiful data have improved our understanding of interhemispheric transport and hemispheric emission gradients.
REPLY: We presume this refers to just the CH_3CCl_3 gradient data, but we would prefer to generalize to include other gases. The use of these gradients to determine regional to hemispheric trace gas emissions is already well covered in sections 3.4-3.6 and 4.7. We add brief text and references in section 3.7, on the use of these gradients for CFCs, etc. to calibrate the 12-box model transport, and in section 3.4, on the way CH_3CCl_3 elucidates the role of ENSO in modulating interhemispheric exchange.
REVISIONS: We replace the second sentence in section 3.7 with: "Therefore, 2D models have been widely used to analyse long-term trends in AGAGE data. The AGAGE 12-box model (Cunnold et al.,1994; Prinn et al., 2001, 2005; Rigby et al., 2013, 2014) uses transport parameters that have been "tuned" using AGAGE observations of trace gas trends and latitudinal gradients (e.g. Cunnold et al., 1994; Rigby et al., 2013) so that the model can simulate monthly mean observations at background AGAGE stations with pollution events removed. From these simulations, multi-decadal AGAGE time series have been used to estimate trace gas global emissions and atmospheric lifetimes.", and add at the end of section 3.7: "A more simplified "3-box" 2D model has also been employed to simultaneously estimate CH_3CCl_3 and CH_4 lifetimes and emissions using AGAGE observations of interhemispheric differences and growth rates (Rigby et al., 2017)." At the end of the first paragraph of section 3.4 we add: "The CH_3CC1_3 seasonal cycle at the tropical South Pacific station (Samoa) showed remarkable sensitivity to the E1 Nino-Southern Oscillation (ENSO), which was attributed to modulation of cross-equatorial transport during the northern hemisphere winter by the inter-annually varying

upper tropospheric winds in the equatorial Pacific; this was a previously unappreciated aspect of tropical atmospheric
tracer transport (Prinn et al, 1992).” In the Reference section, we add:
“Prinn, R.G., D.M. Cunnold, P.G. Simmonds, F.N. Alyea, R. Boldi, A. Crawford, P.J. Fraser, D. Gutzler, D.E. Hartley, R. Rosen, and R. Rasmussen, Global average concentration and trend for hydroxyl radicals deduced from ALE/GAGE trichloroethane (methyl chloroform) data for 1978–1990, *J. Geophys. Res.*, **97**, 2445–2461, 1992.”

24/51: …used for estimating regional ….
REPLY: Thanks.
REVISIONS: We add “estimating” before “regional”.

25/17-37: Please clarify what exactly the AGAGE data policy is. From reading this section I get the impression that data are not publicly released until AGAGE scientists have written their own papers building on these data. This would really deviate from data policies that are stipulated by major (US) funding agencies that now require data products from federally funded projects to be released sooner, and within a fixed time frame no matter if and when the Principal Investigators have been able to publish their own papers by then.
REPLY: Because AGAGE is an international research not operational endeavor and because data validation for some gases can sometimes take longer than others, there is not a strict timetable between data acquisition and data submission. For example, an essential step in data validation involves calibration of the tanks by SIO, both before and after their use at the AGAGE stations, to check for drifts in all of the measured species; this can be time consuming. We add text expressing our general guidelines for data transfer.
REVISIONS: We add to section 5: “Data files for measurements are updated and archived at approximately six-month intervals, following quality-control reviews before and at the semi-annual meetings of the AGAGE team. For scientific credibility, we do not submit data on new gases until at least one peer-reviewed AGAGE paper on that new gas has appeared. Because AGAGE is an international research (not operational) endeavor and because data validation for some gases can sometimes take longer than others, there is not a strict timetable between data acquisition and data submission, but generally we aim to archive data 12-18 months after acquisition.”

25/33: …these CO2 data to the

REPLY: They pass on the AGAGE data for all trace gases, not just CO_2.