Interactive comment 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

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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.

General comments ———————-

1/The AGAGE group made the choice to report both measurement and modelling activ-

ities. 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 : âĂć 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 if them but it should be mentioned in the paper that other approaches exit (ex : variationnal approaches not mentioned, OH results from CTMs & CCMs not mentioned, . . .) which may give different results (see also some specific comments) âĂć There is apparently no database associated to the model results reported here. Then is it in the editorial policy of ESSD to report them ? âĂć If maintained, these sections may be a bit more synthetized as it is looks like catalogues (maybe through tables of references per gas) ?

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.

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) ?

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.

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.

Specific comments ——————

âĂć 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. âĂć 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. âĂć P3-L10 : "Affiliate stations use similar but not identical cryogenic pre-concentration GC-MS systems " : what are the main impacts of such differences ? âĂć P3-l22 :" typical measurement precisions Âż : 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 âĂć 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. âĂć Table 1 : Can you comment shortly in the text on the range of precisions form <1% up to 10% for some compounds ? âĂć P5-l12 : how do you attribute local pollution events (red dots on figure 2) ? From Backtrajectories ? Wind direction & speed at the station ? maybe refer to section 3.1 âĂć P6-l11-13 : The European ICOS is not mentioned here. What are the relation between ICOS and AGAGE ? âĂć 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 ? âĂć 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. âĂć P11-l21 : "a precision of at least 0.3 per mil (‰" for individual measurement ? or aftert averaging ? How long ? âĂć P12 : do the GCWerks package keep track of raw data can eventually correct the full timeseries back in time if necessary ? âĂć P13 : The question of comparison of scales and eventual drifts and corrections between networks is an issue for me and diserves more attention. A table ? Do comparisons between networks change with time ? also treated in 3.2. It might be more clear to

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regroup this part with 3.2 or at least refer here to 3.2. âĂć P14-section 2.9 : do these canisters drift with time for some species ? Has it been investigated ? âĂć P15-l1-2 : please provide a more detailed description of the algorithm (1-2 sentences) ? Does it do a frequency analysis ? change magnitudes ? âĂć Section 3.2 : No mention is made of the European ICOS network. Please provide at leat a strategy of comparison if none is done at the moment as ICOS will structure EU GHG observations in the future. âĂć P15-l47-48: a box model is not a CTM for me as transport is treated crudely, but more a conceptual model. Please rephrase. âĂć 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. âĂć P16 : what about the covariances in the R matrix ? âĂć P16-l4-5? Isn't P should be Pprior in this formula ? âĂć 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). Exemples : Houweling or Saunois papers for methane, Montzka or Krol papers for CH3CCl3, ... âĂć P16-l30 : for the aggregation error please quote Kaminski et al., 2001. âĂć P16 : did you address of diagonal terms of P and R (error covariances ?) âĂć P17, section 3.5 : Why moving from Hysplit to LPDMs ? Please provide minimum differences/improvements. âĂć P17-l43 : "unless an adjoint model of the CTM is available" : please provide a reference, e.g. Chevallier et al., 2005 ot Meirink et al., 2008. âĂć 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 bacause of its low computing cost. Please rephrase. âĂć 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 inversion infer similar results ? âĂć Section 4.2 : I suggest to change the order of the points

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to put first direct results from AGAGE observations (2, 4, 5, 6) and then the analyses (1, 3?). âĂć P20-l14 : GHG → anthopogenic GHG. âĂć P20-l21 : idem for N2O âĂć P21-l4 : how many lead authors & contributing authors ? may be worth précising this. âĂć P21-l18 : what GWP has been used to get CO2-equivalent emissions ? is it for a 100-year horizon ? Please precise. âĂć P23-l10-11 : for HCHC22 Fortems-Cheiney et al. (2013) can be quoted, as can be Fortems-Cheiney et al. (2015) for HFC-134a âĂć 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) âĂć P24-l9, . . . and tranport model errors âĂć P24-l15 : "to help resolve this issue" may be replace by : "to quantify such unreported emissions" âĂć P24-l23 : "Recent" : ambiguous here as 2008 was ten years ago. Please be more precise.

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