Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2021-793-RC1, 2021

Comment on acp-2021-793

Anonymous Referee #1

Referee comment on "Continental-scale contributions to the global CFC-11 emission increase between 2012 and 2017" by Lei Hu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-793-RC1, 2021

Review of the article: "Continental-scale contributions to the global CFC-11 emission increase between 2012 and 2017" by Hu et al.

The manuscripts presents inverse modelling results for emissions of CFC-11 at a continental scale in order to better determine the source of a prior-reported global emissions increase. It uses measurements from two aircraft campaigns (for two different periods) and measurements from NOAA's high frequency and flask network under an emsemble of measurement and a priori emission scenarios. This manuscript will be well received due to the mystery of the source of increased CFC-11 emissions, likely due to unreported production. My only major concern is that the prior uncertainties are not clearly communicated, and as such it is impossible to deduce whether the inversion is actually providing any new information. This must be seen before conclusions can be judged. I hope this is just a matter of clarification, and I hope to see the eventual publication of the manuscript.

Below are a number of suggestions for revisions to improve the manuscript, followed by technical comments, before the paper is suitable for publication.

- Section 2.2.3: This section can be cut down drastically. Rather than providing information on all approaches tried, simply state the one approach taken in the work. It is useful to hear the alternatives used, but is much better suited to relegation to supporting information.
- Section 2.2.4: The use of e.g. "population_87_NA" is confusing as the reader is provided with no information of what that is. They also do not appear anywhere else in the text (perhaps save the definitions in an expansion of the caption for Fig 2). This whole section would be much clearer without the use of the "population_XX" terminology. Further, "prior emissions" are a distribution, not a single value. What you are referring to here is the prior mean(/median/mode as it's Gaussian). 'A priori' may be another useful term if defined. This applied throughout the manuscript. There is also no information on the uncertainty given in your prior distribution, which makes it impossible to understand the information content/uncertainty reduction provided by the

measurements. I suggest providing this information in Table 1 and Fig 6/7.

- Lines 366-375: It is stated that r^2 is less than 0.1. It's unclear which definition of r^2 is used here: there are multiple, some of which can yield negative r^2. As such, it's not clear whether there is in fact negative correlation between the regions.
- It is not fully clear to me how the emissions and uncertainties are derived from the ensembles. In Table 1, I assume that the 'range of the best estimate' refers to the range in the MAP solution across the ensemble members? I don't understand the definition of the 2nd set of uncertainties – is this the range of the lower/upper 2-sigma values? This needs a clearer explanation in the text. It's not currently clear to me whether the posterior uncertainty for a given ensemble member is greater or smaller than the ranges presented.

Technical comments:

Abstract: ACP guidelines state that there should be no references in the abstract unless urgently required. I do not believe that to be the case here. Acronyms defined in the abstract should be defined again in the main text.

Line 17: "early detection" is subjective. Many wouldn't consider 8 years later as early detection. Please amend.

Line 19: "parties to the MP"

Line 60: "policy makers and industrial experts"

Line 66: What is the definition of 'eastern mainland China'?

Line 79: NOAA should be defined

Line 81: What's the definition of "regional-scale" here?

Line 86: An outline of the structure of the paper would be useful here.

Line 91: Were the exact authors included in all studies defined as 'ours'? If not, delete 'our'.

Line 93: Upwind from the measurement location

Line 94: Change "is footprints or" to ", termed footprints, are".

Line 96: State that these models are HYSPLIT and WRF-STILT.

Line 97: Pedantic, but Bayesian inversions *always* require prior probabilities, regardless of the constraints.

Line 104: Here z is an enhancement. Please reference here the later section on how you derive the enhancement from the measurement?.

Line 109: Again pedantic, but "maximum likelihood estimation" is by definition non-Bayesian. If Bayesian, you are finding the maximum a posteriori (MAP) solution.

Line 130: Be consistent with the use of hyphenation of in situ/in-situ (I would remove it).

Line 155: Change ‘our’ to ‘the’

Line 156: relative to what? Best to give an approximate distance for how [far] away the sampling is from emissions.

Line 219: How long were they run back in time?

Line 344: Change “described above” to “described in Sect. 2”.

Line 363: It seems incorrect to me that 16 ± 10 Gg/yr then only has a percentage uncertainty of 80-90% of the global total.

Line 411: Change “given we’ve already know[n]” to “given it is known”.

Line 431: How are you defining the ‘best estimates’?