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Comment on acp-2021-633

Anonymous Referee #1

Referee comment on "Swiss halocarbon emissions for 2019 to 2020 assessed from regional atmospheric observations" by Dominique Rust et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-633-RC1>, 2021

Review of:

Swiss halocarbon emissions for 2019 to 2020 assessed from regional atmospheric observations

Dominique Rust et al.

This paper describes Swiss emissions of a large number of greenhouse and ozone-depleting gases from two top-down methods and compares these results with those officially reported or derived from Jungfraujoch observations. The new observations from Beromunster, Switzerland are introduced and form the main basis for the paper, although other observation stations are used in the inverse modelling.

General comment

For quite a few gases there are very significant differences between the TRM and the BI methods. It is therefore a reasonable question to ask whether the times of good agreement are just fortuitous. The key question that has to be addressed is why there is there good agreement sometimes and poor on other occasions. This does undermine the credibility of what is presented if this is not addressed. Which method do the authors believe is the better method?

Substantive Points

P.7, L225: "for a larger emitting region and over an increased time period" – Larger than what and increased relative to what? Please can this sentence be clarified.

P.8, L250: "specific sigma factors, i.e. 1, 1.5, and 2" – Are these not better described as multiplication factors? If I understand correctly this means that when the factor is 1, more points are considered 'polluted' as they are above the baseline + (baseline uncertainty)? Please can this be made clearer? In fact if table S4 (there is no Supplement 4.1) had extra columns the values for factors of 1 and 2 could be included making this very clear.

P.8, L.272: "were added to the" – This was unclear to me, you cannot just add 2%, say, to both elements? You could add 2% to the top and subtract 2% from the bottom and vice

versa, to give a range. Please can this be clarified.

P.9, L.294: "0.2° by 0.2° in the Alpine area and 1° by 1° elsewhere" – How big is the Alpine area? Also 1 degree (110km) seems very coarse for modelling coastal Mace Head and Tacolneston (~50km from the coast) sites. Has this been demonstrated to be sufficient? What impact would having 0.2 degrees everywhere make?

P.9, L.296: "for 4 and 10 days" – How far do the particles travel in these time scales on average and as a minimum? The length of the simulation will naturally affect the residence time percentages used in the TRM? Also it seems reasonable that all the particles will have clearly left the Swiss area within 4 days but it is not clear the background will be fully mixed, i.e. a source just beyond the 4 days may still be very discernible. Can the authors be confident this is negligible?

P.9, L.315: "Daily mean values" – In previous work that I have seen using this method, 3-hourly averaging times were used. Is there a specific reason why the observations were averaged into daily values? It seems odd to lose all this extra information, I think some justification of this is necessary.

P9. L.319: "12.0o W to 21.1o E and 36.0o S to 57.5 o N" – Given that you follow the particles for only 4 days from the Swiss sites then it seems reasonable that many of the released particles will not have left your inversion domain within the 4 days. For example a direct southerly wind of 3 m/s consistently for 4 days is insufficient to move a particle from the southern edge (36deg N) to Beromunster (47degN). How is this accounted for? Also I assume it is a typo as you have written 36 degrees South, should this not be 36 degN?

P10. L351: "The HFOs were treated as inert for the inversions, assuming that the transport times from emission sources to BRM are sufficiently small to avoid larger chemical losses" – Given the very short lifetimes of some of the HFOs (single digit days in summer for HFO-1234yf), I think the authors need to quantify the potential error here and also the bias between summer and winter.

P10. L354: "HFC-23, SF6, and PFC-14" – The emissions of PFC-14 from Al production and HFC-23 as an industrial bi-product will not be population based. So even though the final statistics maybe improved using a population-based prior, is the use of such a prior reasonable or in any other way justifiable? If little is assumed known about the distribution of emissions is not using a 'flat' prior more reasonable? Also I assume the authors meant 'flat, land-based' prior rather than flat across the inversion domain – please clarify.

P.18, L548: "The results compare well to the Jungfrauoch and the inventory values" – I am not convinced that this is the case for all of these gases, e.g. HFC-145fa and HFC-227ea and HFC-23. A similar comment can be made about PFC-14.

P.19, L.560: Why is this gas so different across the methods?

Minor Points

P.2, L.63 : "it is used as aerosol propellant" – Insert "an"

P.2, L.63: "and as foam blowing agent" – Insert "a"

P.3, L82: "HFO-1234yf is currently applied as refrigerant" – Insert "a"

P.3,L85: "and as foam blowing agent and propellant" – Insert "a"

P.4, L.127: "which are constantly monitored within the AGAGE network" – Remove the word "constantly", they are measured at high-frequency not constantly.

P.4, L.148: "industrially most active region of Switzerland" – Insert "the"

P.5, L163: "from the latter on a" – Suggest changing to "from this area on a "

P.7, L226: "are at a significant distance" – Please remind the reader of distance to the nearest large town e.g. more than 20km.

P.8, L.255: "were weighed accordingly" – Change to "weighted"

P.8, L.255: "to result in a CO emission" – Add the word 'Swiss'

P.8, L.264: "in Supplement 4.2." – Better to say Supplement Fig. S2 as 4.2 doesn't exist.

P.8, L.266: "pollution events were summed up" – Please remind the reader that the baseline has been removed to estimate a pollution event.

P.8, L.280: "(Supplement 4.3)" – Supplement Table S4, 4.3 does not exist

P.8, L.282: Should section 2.4 come before 2.3 as these simulations are used for the residence times?

P.9, L.304: "Next to total receptor mole fractions" – Not sure I understand the use of the phrase 'Next to', what does it mean?

P9. L323: "δ□□□δ□□□ to the observations" – In the previous paragraph the observations were referred to as 'y', please change for consistency.

P9. L325: "model-observation uncertainty" – Adding the word 'respectively' at the end of the sentence will help the reader.

P9. L325: I think it would be helpful to the reader to very briefly describe the spatial and temporal covariances used to construct B and R rather than rely on them reading another paper.

P10. L333: "(δ□□□ 0 , δ□□δ□□¥)" – Here the O is a superscript, previously it was a subscript.

P10, L349: "countries of the inversion domain" – Consider changing 'of' to 'in'

P10. L359: "chi index" – I think this term needs greater explanation with an appropriate reference.

P10. L367: "different substances was evaluated" – please quantify how this was done.

P.12, L.381: "atmospheric concentrations" – consider changing to 'atmospheric mole fraction'

P.13, L404: "small emissions in Switzerland" – add HFC-152a to this for clarity.

P.13, L413: "there was no notable number of" – consider changing to 'there were no notable'

P.13, L.415: "emitted as unwanted" – – consider changing to 'emitted as an unwanted'

P.13, L.417: "major fraction of the highest events" – please quantify

P.17, L.499: "mostly used in refrigeration" – 'mostly used as a refrigerant'

P.17, L.502: "are invariably higher" – remove the word 'invariant'

P.18, L.531: "third highest emissions" – 'third highest Beromünster emission estimate'