

Atmos. Chem. Phys. Discuss., author comment AC2
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Reply on RC2

Eric Förster et al.

Author comment on "Chemical and dynamical identification of emission outflows during the HALO campaign EMeRGe in Europe and Asia" by Eric Förster et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-455-AC2>, 2022

Author response on anonymous referee #2 (R2), acp-2022-455

Review of Chemical and dynamical identification of emission outflows during the HALO campaign EMeRGe in Europe and Asia, by Förster et al.

This manuscript details the use of observations of a few specific VOCs made during two HALO flight deployments to identify the influence of major source types by using threshold values of compounds to identify influence of biomass burning emissions, anthropogenic emissions, biogenic emissions, or combinations of the above. The authors then use back trajectory modeling to attribute and quantify these various contributions back to specific regions.

Answer: First of all, we would like to thank R2 for carefully reading the manuscript and for the generally positive, helpful comments as well as the detailed technical corrections. We have followed all suggestions for change and have modified the manuscript and Supplementary Information accordingly. Below we provide point by point responses to the *comments* with given lines of changes (if applicable) and highlighted the corresponding changes made in the manuscript (supplement file) in blue. Changes corresponding to comments of R1 are highlighted in yellow.

Author response

The presentation of the research is a little tedious to get through, but in general, it seems like a reasonable methodology for broadly assessing air masses sampled at varying distances from their likely sources. I have a few issues with the exact approach, however. One major issue I have is that the authors use in their back trajectory modeling to the PBL is that it discounts biomass burning injection height, and lofting due to convection, which can be significant over the Tibetan Plateau due to the Asian Monsoon. The authors should address this possible contribution and implications in their in their work.

Answer:

We agree that lofting by convection is an export mechanism of emissions, especially in the

tropics. Since we i) only wanted to set up a simple model and ii) most of the probed emissions emanate from mid-latitude sources (here low-level advection dominates according to Folberth et al., 2015), we have decided to neglect lofting by convection. We added a paragraph (lines 292-296) describing this point.

We also agree that over the Tibetan Plateau convection is a significant transport mechanism. However, in the case of EMeRGe-Asia, the effect of the Tibetan Plateau can be neglected because i) we measured in an inter-monsoon period (March/April) and ii) there are nearly no emissions of BB and anthropogenic sources over this region that could be transported. So the impact would be negligible.

A biomass burning injection height is not applicable here because we only consider anthropogenic CO in the trajectory model. We added a clarification in the introduction of Sect. 3.2.2. (lines 290-291).

Secondly, the authors appear to put far more emphasis on the value of the back trajectory modeling than in the VOC tracers that they are studying.

Answer: Due to the multiple targets of EMeRGe and the large-scale probing, the dynamical identification of pollution outflows (back trajectory modeling) is an important part of the campaign analysis to determine/estimate the overall source contributions during EMeRGe. The VOC tracers alone are a helpful tool to characterise air masses, but in the case of EMeRGe the larger emphasis on the modelling was necessary to identify the origin of the probed air masses, and hence to characterise specific pollution outflows of target areas.

Lastly, due to the nature of aircraft campaigns, there is an inherent sampling bias to air masses from specific source regions, and that should also be addressed in the paper.

Answer: The issue of a sampling bias is now addressed in the manuscript (lines 137-138). We want also to emphasise that we did not make any conclusions, which can be invalidated due to this sampling bias.

Some more specific comments follow, with a large number of technical corrections below.

Answer: We thank R2 for the detailed specific comments, which have all been implemented.

Line 37 – consider using “rural areas” instead of “countryside”, and perhaps consider that in many countries there is a suburban interface, and identify whether that is “urban” or “countryside/rural” in the UN statistics.

Answer: Changed to rural areas (line 37). The UN (United Nations, Department of Economic and Social Affairs, Population Division, 2018) uses the three concepts “city proper”, “urban agglomeration” and “metropolitan area”:

“... “city proper”, describes a city according to an administrative boundary. A second

approach, termed the "urban agglomeration", considers the extent of the contiguous urban area, or built-up area, to delineate the city's boundaries. A third concept of the city, the "metropolitan area", defines its boundaries according to the degree of economic and social interconnectedness of nearby areas, identified by interlinked commerce or commuting patterns, for example."

We would see the suburban interface as additional area between urban agglomeration and metropolitan area, which is included by the UN for 10 % of the evaluated cities:

"The 2018 revision of World Urbanization Prospects (WUP) endeavoured wherever possible, given available data, to adhere to the "urban agglomeration" concept of cities. Often, however, in order to compile a series of population estimates that was consistent for a city over time, the "city proper" or "metropolitan area" concepts were used instead. Of the 1,860 cities with at least 300,000 inhabitants in 2018 included in WUP, 55 per cent follow the "urban agglomeration" statistical concept, 35 per cent follow the "city proper" concept and the remaining 10 per cent refer to "metropolitan". (United Nations, Department of Economic and Social Affairs, Population Division, 2018)

Line 75 – "(and that provides only one time information based on its lifetime..." is not communicating what the authors are trying to say.

Answer: The text was adapted to "Such an analysis is advantageous compared to the conventional approach of using a single tracer like carbon monoxide (CO) with its rather long lifetime (~2 months) and multiple sources (anthropogenic and biomass burning)." (Lines 76-77)

Lines 115-116 – "some 100 km" – from what?,

Answer: Changed to "100 km downstream of targeted source regions". (line 120)

and on the next line, perhaps "source" instead of "target"? since the regions are where the emissions are coming from, not heading to.

Answer: Changed to "targeted source region". (line 120)

Also, "concept is mirrored by"... mirrored doesn't seem like the right word here.

Answer: Changed to "reflected". (line 121)

Also, one might argue that at 100 km, emissions are not exactly fresh.

Answer: Changed to "recently emitted". (line 120)

The other reviewer makes very good points about emissions vs. characterizing source

concentrations, so I won't belabor that point, but I will echo that it needs to be addressed throughout.

Answer: We agree that the term "emission" was used in two ways throughout the manuscript: First as term for a general description of "release of trace gases" from various sources, e.g. biogenic or anthropogenic, without a quantitative statement. And second, as modeled CO emission uptakes, with a quantitative indication.

For the first case (the measurements), we mostly changed the term "emission" to "pollution" or "composition" throughout the manuscript as suggested by R1. We added also a short paragraph defining pollution as emission impact (lines).

Since the modelled emission uptake is given as a mass, we kept the term for the modelling of the anthropogenic CO uptake. However, we tried to be more precisely and using now mostly the terms "emission uptake" or "modelled emission" throughout the modelling discussion.

Line 162 – "to the scientific needs" requires more explanation.

Answer: We added a more detailed explanation (lines 173-174): "... scientific needs, e.g. customisable duty cycle of measurements/background detection due to meteorological conditions/ conditions of aircraft campaign and full access to all operating parameters."

Table 3 – is there a reason why some columns are italicized and some aren't? This isn't clear.

Answer: We thank R2 for the note. Originally, the italicized columns should designate the source signatures I with the corresponding VOC enhancements (or no enhancement) of acetonitrile and benzene used for the definition. We have removed italics from all columns to avoid confusion.

Lines 223-225 – it is also not clear to me how nine consecutive 6-s measurements can be reasonably interpolated to 1-s data.

Answer: We thank R2 for the remark. Actually, we did not interpolate the VOC measurements itself to 1s, but extended the assignment of source signatures (based on the measurements of three VOCs) to the general measurement frequency of 1s. We rephrased the text in the manuscript (lines 240-242) and in the supplement:

"Due to the PTR-MS duty cycle (consecutive integration of nine VOCs with ~6 s each), we assign the identified source signatures to the general measurement frequency of one second to enlarge the data coverage (see supplementary Sect. S4 for detailed description)."

Line 251 – replace reddish colours with "red", as the high emissions are just one colour of red.

Answer: Text was adapted (line 278).

Figure 2 – these plots are very difficult to read, and are far too busy. Perhaps reduce the intensity of the background colours so that the text, flight paths, etc. can contrast against the background.

Answer: The colour scheme of Figure 2 was adapted.

Lines 562-563 – In 19% and 34% of what are negligible contributions inferred? Be specific.

Answer: Text was adapted (lines 596-597): Moreover, in 19% (EMeRGe-Europe) and 34% (EMeRGe-Asia) of the total flight time small/negligible contributions are inferred and not considered.

Technical corrections:

Answer: We thank R2 for the detailed technical corrections which have all been implemented.

Throughout A: "air-mass" or "air-masses" should be changed to "air mass" or "air masses".

Throughout B: remove spaces before % signs (e.g., line 28: "20 %" should be "20%".)

Throughout C: refer to style guide on referencing sections, specifically "The abbreviation "Sect." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence."

Throughout D: "back trajectories" doesn't have a hyphen.

Throughout E. Be consistent with "up-take" vs. "uptakes" – the latter of which is correct (e.g., Table S4)

Line 38 – "the majority of megacities are still..."

Line 39 – what is meant by "extension"? Maybe not the right word.

Answer: The spatial extent of a city. Changed to "dimension". (line 39)

Line 48 – "... focused, e.g., on emissions..." (add commas)

Line 50 – there appears to be an extra space after "2008-2011"

Line 54 – "(Andrés Hernández et al., 2000 and references therein)" (remove inner parentheses)

Line 62 – "refer to Andrés Hernández et al. (2000)." (no comma)

Line 83 – "joined" should be "joint"

Line 119 – define ERA5

Line 123 – instead of "local burning", use "local fire".

Line 125 – for simplicity, use "chemical age, i.e., chemically old..." rather than "that is"

Line 129 – "starting from" (not form)

Line 135 – "10-day FLEXTRA", "back trajectories" (no hyphen).

Line 146 – remove the space before H3O+.

Line 151 – "vast majority of VOCs in the atmosphere are" (not is)

Line 152 – delete "further"

Line 156 – "nighttime"

Line 157 – "lifetimes"

Line 162 – "allows adaptations"

Line 164 – limit of detection = LOD, lower detection limit = LDL? Maybe use "lower limit of detection (LOD)"? Also, "in the pptV range". (no hyphen)

Answer: Changed to "limit of detection (LOD) in the lower pptV range". (line 175-176)

Lines 168-170 – "tropospheric lifetime t ", and again, "lower limit of detection (LOD..." change "and the up-to four atmospheric main sources" to "... and the up to top four main atmospheric sources."

Line 178 – maybe "as explained below."

Line 181 – "lifetime"

Lines 189-190 – "please see details in the supplement" – perhaps instead refer to a specific supplement

Line 205 – "... VOC tracers are..."

Line 218 – "lifetimes"

Line 225 – again, perhaps point to a specific section in the Supplement.

Line 235 – "10-day back trajectories..."

Line 236 – “the time step...” (no hyphen)

Line 251 – replace reddish colours with “red”, as the high emissions are just one colour of red.

Line 265 – delete “Earth’s” (it is redundant.) and perhaps use “trajectory air parcel” instead of air mass.

Line 266 – recommend changing to “... emissions rates at that location.”

Line 272 – replace “exemplary” with “example”

Line 276 – immerses is not the right word here. Even “dips” would be better, or “drops”, “descends”, likely best.

Line 291 – define AMTEX

Line 304 – “... values listed in Table S2).” (S indicates supplement, so “in the supplement” is unnecessary).

Line 357 – “Figures 5 and 6...”

Line 365 – “the blue dot” is very difficult to see. Again – I recommend if possible having a less intense colour scheme for the background colour bar information, so that the details on top can be seen. Same comment for Figure 6.

Answer: The colour scheme of Figure 5 and 6 has been adapted accordingly to Figure 2.

Line 381 – “some hot spots such as the...”, also include a comma after Madrid.

Line 385 – “More regions have likely...” – begs the question “more than what”? Be specific.

Answer: Adapted the text: “Compared to EMERGe-Europa, more regions have likely contributed ... ” (line 419)

Line 397 – “Coordinates are shown in Table S3.” Is sufficient.

Line 402 – “Figures 7 and 8...”, also, Line 405 – “Figs. 7 and 8...”

Line 411 – BeNeRuhr needs to be defined. And consider whether two short forms (BNR as well) might add confusion.

Answer: Now only BNR is used as abbreviation throughout the manuscript.

The text in Figures 7 and 8 in the blue boxes is really difficult to read. Also, for both captions, “(plain) is not a great description. Perhaps use “the last row in each section”, or

"the uncoloured row".

Answer: The colour scheme of Figure 7 and 8 has been adapted accordingly to Figure 2, 5 and 6. "Plain" was changed to "uncoloured rows".

Line 530 – "20 July 2017".

Line 604 – "fresh biomass burning" or "fresh fires"

Supplement

Line 3 and Table S1 – "Takeoff" and "Departure" are the same thing. I think you mean Takeoff and Landing, or Departure and Arrival. Just a note, as well – this may be the convention for HALO flights, but using ICARTT nomenclature, the flight date is always the UTC departure/take-off date, regardless of how close it is to midnight, UTC.

Answer: We changed the dates to the departure/take-off date. This is also the convention for HALO flights; however, we originally used the two "shifted" dates internally.

Line 10 – "measurement noise by"

Line 17 – "4 April 2018"

Line 19 – Instead of writing "To Section 3.1.2, give the various sections in the supplement Section numbers (e.g., Sect. S1, Sect. S2.4, etc.), which will make reader navigation between the main manuscript and the supplement much more straightforward.

Line 42 – "Same applies for isoprene." Is not a complete sentence.

Answer: Added isoprene to the first sentence ("...", where the tracers acetonitrile, isoprene and benzene are measured ..."). (line 33)

Line 43 – "This assumption..." or "These assumptions..."

Line 45 – "as an additional tracer..."

Line 48 – this is far too much prose for a Table title. Move most of this to a text paragraph or to table footnotes. Also, the last line "This figure is a supplement..." This isn't a figure - it's a table. Also, I recommend only putting "Table X" or "Figure X" in bold, and not the entire title or caption.

Answer: The table caption has been shortened and/or moved to a footnote (we shortened

the caption of Figure 4, 10 and 11 in the main text accordingly). The entire caption is bold by default in the ACP MS Word template. In a published version, it will be only the label and not the whole caption, which, we agree, is indeed better.

Line 58 – "MPCs (in italics) with coordinates..." or "MPCs (italicized) with coordinates..."

Lines 64 and 66 – delete "... in the main document." (it is unneeded, as sections and tables that don't begin with S are by definition in the main documents.

References

Folberth, G. A., Butler, T. M., Collins, W. J. and Rumbold, S. T.: Megacities and climate change - A brief overview, *Environ. Pollut.*, 203, 235–242, doi:10.1016/j.envpol.2014.09.004, 2015.

United Nations, Department of Economic and Social Affairs, Population Division: The World's Cities in 2018, *World's Cities 2018—Data Bookl.* (ST/ESA/ SER.A/417), 34, 2018.

Please also note the supplement to this comment:

<https://acp.copernicus.org/preprints/acp-2022-455/acp-2022-455-AC2-supplement.pdf>