

Review of “Detection of mixing and precipitation scavenging effects on biomass burning aerosols using total water heavy isotope ratios during ORACLES” by Henze et al.

This paper presents very innovative research on the use of water isotopes to assess the impact of different atmospheric processes on biomass burning aerosols using observations from the NASA ORACLES field campaign. I enjoyed reading this very interesting manuscript! I have no major scientific comment, the work is of high quality, but I would strongly recommend the authors to refine the writing and presentation of their results (figures quality). Given the wide range of tools and expertise that is combined in this publication, more clarity in the writing is essential to make these valuable findings more accessible for both isotope and aerosol experts. My minor comments, which you'll find below, as well as a small number of technical points listed at the end of my review will hopefully help the authors to refine the presentation of their work.

Major comments: improve the readability of the text and the reader guidance as well as the quality of the figures.

Minor comments:

- 1) Title: with “mixing” it is not entirely clear what you mean, would it make sense to point out here already the dry vs. moist convective mixing pathway of biomass burning aerosols (BBA) into the free troposphere (FT) that you are addressing in the paper?
- 2) L 11-12: at this stage it is not clear to me that you mean dilution by turbulent mixing in the free troposphere. Maybe it would help me if you wrote “precipitation scavenging” and “dilution due to mixing”. And in the next sentence, I would keep the same order of processes as in the first sentence: precipitation processes versus mixing.
- 3) L. 15: “Air... is distinct, and can be treated as separate analyses...” After reading the full paper I now understand what you mean but without knowing the paper this sentence sounds a bit mysterious. In which respect are FT and MBL air masses different?
- 4) L. 15: since you don't use MBL again in the abstract you don't need this abbreviation there, only in the main text.
- 5) L. 16: remove “to”
- 6) L. 17: “assess of...” I don't understand this sentence, something went wrong with the phrasing here. Maybe you mean “are used to assess the relevance of the air parcels' precipitation history”?
- 7) L. 19: make clear that MERRA-2 is a global reanalysis dataset
- 8) L. 28: CCN is not defined in the abstract
- 9) L. 31: Again make clear that you mean dilution by mixing: “signature of dilution effects by air mass mixing in the free troposphere”. In my first reading I thought you mean dilution of heavy isotope concentrations by rain out underway. I didn't immediately think of mixing. But maybe making this clear at line 12 is enough. Of course, when reading your paper, one wonders what process exactly leads to dilution in the free troposphere. The turbulence is likely very limited except in the environment of clouds or in regions with strong vertical or horizontal wind shear...

10) L. 41: "particularly after again"? There seems to be part of the sentence missing here.

11) L. 44: "...quantifying processes... is of importance for climate models."

12) L. 54: maybe "compared to the reduction..."

13) L. 54: this is the first time the term dilution comes up in the main text, make clear that you mean dilution by mixing through turbulence and convection.

14) L. 57: how about first saying: "they provide information on the relative importance of the history of moist atmospheric processes such as mixing..."

15) L. 60: "While isotope ratio information to date has shown promise...". This is true but a bit unspecific.

16) L. 62: "... for the advantages to be realized". Maybe "to take full advantage of the tracer capacity of water isotopes and their use for assessing aerosol cycling in the atmosphere".

17) L. 67: "The isotopic compositions for water D/H" is a pleonasm. Maybe "the isotope composition of water" or the "concentration of HDO"?

18) L. 70-71: "In the case of the FT, regional aerosol and moisture transport are connected and characterized" what does "connected" mean exactly? That they are considered together?

19) L. 82: How are the relative importance of evapotranspiration, dry versus moist convection etc. constrained? This needs to be explicitly clarified and the (q,dD) phase space is so essential for your paper that it needs to be introduced in the introduction.

20) L. 69-90: this whole section is a bit unspectacularly written and sounds more like a casual report or list of things that were done in this paper (e.g. "Next,... is evaluated."), but in a way that is not rising the readers curiosity and attention. This paragraph is absolutely key and it is very cool! I think the authors could invest in building up some tension here, maybe listing the research questions, rising the reader's curiosity.

21) L. 88: A new paragraph could be started at "Section 2 covers...".

22) L. 89: "Section 3 presents a brief **analysis** of the sub-cloud as a starting point for contrasting the **analysis** of the FT **analysis** of precipitation histories and scavenging". Please rephrase.

23) L. 91: "precipitation metrics", not sure what you mean here, the ones derived in Section 3?

24) L. 106: "Flights were typically every 2-3 days, which 7-9 hours in duration and...".

25) L. 114: I would leave away "Measurements of" and just write "Potential temperature...".

26) L. 146: "Using vertical profiles..." I don't fully understand this sentence. In particular, why there is a logical link between "using RH over liquid water as a flag for the CL" and "profiles through CLs with broken clouds".

27) Section 2.5 first introduce the (q, dD) diagram, it is otherwise a bit difficult to follow and understand what is meant here.

28) L. 171: What do you mean by "subsequent mixtures between them"?

29) L. 187: There are no theoretical lines in Fig. 3, you probably mean Fig. 4.

30) Fig. 3: is there a colormap missing or is it just that you color the points in the total water mixing ratio?

31) L. 201: Rephrase, "for air where moisture from the respective evaporation source..." there is a verb missing or "and" is not needed.

32) L. 203: you probably mean the blue circle in Fig. 4.

33) L. 208-220: In this paragraph one can get lost as an isotope beginner. Guide the reader, what is the aim? Also, it is not entirely clear which variable is measured which is modelled/taken from the reanalysis and what the uncertainty resulting from the use of the MERRA data is.

34) Section 3: the start of the results section is a bit abrupt with a detailed description of Figure 5. The reader could be led a bit more smoothly through the results and its structure. The different results sections come a bit as a surprise.

35) L. 227: Rephrase the first sentence, something went wrong. Maybe remove "of"?

36) L. 230: nice that you refer to previous studies, but this could be done in the intro or the discussion, start with addressing your question. I think Feng et al. 2019 didn't present new observations.

37) Results: From a structural point of view, and given your research questions sketched in the introduction, I am not convinced that starting with the subcloud layer is a good idea. But maybe there is a good reason for this, then state it to make sure the reader knows why he/she is reading this.

38) Results and discussion: Given the complexity of this study, I very much doubt the use of separating the results and discussion section. Now that I made the effort of understanding your very interesting Fig. 5, I would like to learn what it tells me about moist processes in the boundary layer. Until Section 5 (where you discuss the science), I will have forgotten most of what you show in Fig. 5 because I will have had 10 additional complex figures to understand.

39) Fig. 5: When I look at Fig. 5, I don't really see these measurements as fitting particularly well in a mixing framework or along an imaginary mixing line.

40) L252: what is the output frequency of the interpolated air parcel positions? Are hourly 3D wind fields used? The information on the trajectory calculation setup belongs into the methods section.

41) L258: "double that of the SCL" I don't' understand this. What is the CO concentration of the SCL, is it shown somewhere?

42) L262: remove it, "The low-BBA air in 2016 is..."

43) L285ff: Make clear that this assessment with (monthly?) surface precipitation collocated with trajectory information is a first coarse assessment of the possibility of precipitation formation underway. It is not clear if 1) precipitation did really form at the time of the airmass overpass over a given region (timing uncertainty) and 2) if the airmass that you are tracking is really the one involved in precipitation formation, it could be that it is travelling above the cloud (vertical level of precipitation formation uncertainty)

44) L300ff: The sources of water vapour and BBA are likely not the same... This should be critically discussed somewhere. Did the authors look at forest fire observations from remote sensing to identify potential BBA sources?

45) L347ff: I am not sure, I understand why correlations are computed between dD and altitude, and in which context to place the vertical gradients of dD. Here the reader needs help to understand why these analyses are done.

46) Table 3: Was isoCAM evaluated with respect to near-surface observations over tropical Africa? How confident are the authors that this data is reliable? How is the

treatment of ET done in isoCAM? Is there a multilayer soil model to take near-surface soil layer enrichment due to evaporation? Is soil evaporation and plant transpiration treated in a fractionating or non-fractionating way?

- 47) L385: why was this data discarded? It would provide a very interesting control group and reference point in the dD-q phase space for “clean” airmasses with likely important moisture contributions from the MBL.
- 48) Figs. 9 and 10 are complex and thus a bit too small to understand well.
- 49) In Fig. 11 many data points seem to be overlying each other. This makes the figure difficult to read.
- 50) Fig. 11 and 12 and related results: I really like this approach, but I got lost when trying to understand how the convective plume model was exactly set up for these calculations. There are several unconstrained parameters in this model. I think a dedicated methods section on the setup of the convective plume model would really help me to see through the approach.
- 51) To me turning back to the SCL and Fig. 5 now feels like quite a slalom.
- 52) L. 449: CCNs are already introduced as an abbreviation, define it above when first mentioning them in the main text.
- 53) L496: As already mentioned earlier I find this part about the subcloud layer a bit weak. The motivation to assess the dD “for agreement with basic theory” is not very convincing. I like the idea of assessing the horizontal and vertical variability of the dD in the MBL and it would be nice to have 1 or 2 short statements about where this variability comes from process-wise.
- 54) L520ff: I am confused by the reasoning behind the first paragraph of this subsection: why should the high-BBA PDF be linked to the SCL observations? I thought that the hypothesis is that the transport pathway of moisture into the free troposphere is: 1) ET over tropical Africa, 2) dry or moist convective detrainment into the free troposphere, 3) Advection over the South Atlantic. So, if there is a link with the SCL measurements in the MBL that would be by convective downdrafts bringing dry FT air into the SCL, right?
- 55) L558: I can't really see that in Fig. 13, to me it seems that many data points in all three IOPs deviate a lot from the mixing and Rayleigh lines.
- 56) L560ff: I must have missed it but is the MBL defined as the SCL and the CL in your study? Could that be included in the sketch in Fig. 1.
- 57) L568: that's very interesting!
- 58) L611: Repeating the fact that three IOPs over three years in the late winter period August-October with xx number of flights and in total yy hours of measurements were involved would be great.
- 59) L612: in combination with?
- 60) L613: “of moisture contributions from different processes along the atmospheric transport pathways of water vapour (e.g....)”
- 61) L615: “accompanied by cloud formation and precipitation...”
- 62) L625 & L626ff: “This diagnostic partitioning cannot be achieved with humidity measurements alone...” very nice result indeed!!
- 63) L655: It would be fantastic if the paper could end with a more general closure statement of why this combination of isotopes with aerosol data is useful and why research on this combination of observations should be further pursued in the future.

Technical comments:

- The Figures are all very interesting, but the quality (resolution?) is a bit poor. It's a pity, if it stays like this because of their very valuable content.
- There are many instances with excessive spaces, double .. or , or missing commas.
- To make this complex paper (in terms of the number of aspects and processes involved) easier to access to the readership, I would strongly recommend reducing the number of abbreviations. There are some that are clearly needed, but others are used only a few times and could be written out. For example, for the boundary layer there is SCMBL, MBL, PBL... do you really need all of them or could you just abbreviate BL and then write out the specific one you mean? Or for example biomass burning (BB) you don't use so many times, you mainly use BBA, so could you write out "biomass burning" in the few BB instances? I think similar considerations for other abbreviations would be worthwhile to make the paper easier to read.
- Many abbreviations are not properly introduced in the text, they should be written out the first time they appear (PBL, MBL, LCLT, CCN in the abstract).
- Find a consistent term to refer to the (q,dD) phase space, it is sometimes (q,dD) and at other times q, dD. Also, this is such an important tool for the paper, could it be mentioned already explicitly in the abstract?
- All variable names in italics, e.g. R_D (L. 113). RH is sometimes in italics sometimes not (e.g. L. 152 and L. 155). However D as a chemical notation for deuterium should not be in italics.
- Airmass is sometimes written in one word sometimes in two (air mass). The same for end member vs. endmember.
- References mentioned directly in the text like Fiorella et al. (2022) have no comma: E.g. not Fiorella et al., (2022) at L. 316.