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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal source variability of carbonaceous aerosols at the Rwanda climate Observatory" *by* August Andersson et al.

Anonymous Referee #3

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"Seasonal source variability of carbonaceous aerosols at the Rwanda climate Observatory" by Andersson et al.

The manuscript reports results of ground-based observations of carbonaceous aerosol at a mountain site in Rwanda, sub-Saharan Africa. Fine airborne particulate matter (PM2.5) was collected weekly basis on quartz fibre filters, then total carbon, organic carbon, elemental carbon, water-soluble organic carbon contents and their radio and stable isotopes were analyzed. Major inorganic ions (nitrate, sulfate, ammonium, and potassium ions) were also analyzed. Mass balance approach was applied to the isotopic data to identify the origins of the carbonaceous aerosols. The results were also

compared with wild fire information provided by NASA and with back trajectories of air masses arriving at the sampling site using HYSPLIT provided by NOAA to better understand the origins.

Studies of origins of airborne particulate organic matter are one of the hottest themes in atmospheric science, but the origins are complex and not easy to be untangled. Application of isotopes measurements will provide additional dimension in the scientific information, thus, ideal for such studies. I have been aware of the authors' previous work, the dual isotope analysis, and the analysis provides insight into the origins of organic carbons, particularly differentiation between biogenic and fossil fuel origins using 14C. Compared to 14C, the method of 13C fingerprinting is week due to its large variation of the end members, depending on source types. Here, the authors attempted estimation of carbonaceous aerosols derived from C3 and C4 plants, which are known to have discrete compositions in 13C from each other. Papers reporting those fingerprints of organic aerosols are not many (Mkoma et al., 2014, TellusB; Irei et al., EST, 2014). The 13C fingerprinting approach to the carbonaceous aerosols that contain secondary organic aerosols would not be as simple as the authors describe here, but the studied location may be close enough to capture primary carbons.

As I read this manuscript, the analysis was relatively straightforward, and gained results sound reasonable. However, I also had impression that they could go into a fairer evaluation that considers the possible variation in the 13C compositions of endmembers. For example, according to L177-179, they performed Monte Carlo simulation to estimate the uncertainties of fC3, fC4, and ffossil propagated from the variability of fingerprinting 13C compositions. The gained uncertainties were small enough to capture significant values of source contributions and then plotted in Figure 6. At the same time Figure 5 shows wide ranges of the 13C compositions for the end members, C3, C4 plants and fossil fuel. Are those variations reflected to the uncertainties of the source apportionment? Such large variations do not seem to end with the uncertainties in Figure 6. If not, I recommend the authors to work on more objective evaluation

by providing a several scenarios with the combinations of different 13C compositions of the three endmembers, together with the most feasible apportions (probably those currently described in the manuscript). This will raise the quality of this manuscript, I believe. My comments on specific parts of the manuscript are provided below.

Abstract: Remove "(PM2.5)" because the abbreviation is not repeated in the abstract.

L98: Add the manufacturer information for PM2.5 inlet. Please provide the information of sampling frequency and duration.

L151-152: Please provide the model and manufacturer information for AMS and IRMS.

L192: Correct JAA to JJA.

L193-194: Figure 1 does not show distinct plots of wild fires for the JJA and DJF periods. The authors may want to show two plots showing two periods.

L205: Please define "BT".

L214-215: "This variability suggests. . ." needs supporting information (i.e., references).

L221: Replace "-" with commas.

L223: Provide references directly here instead of "Table 1 that refers references.

L224-226: Please explicitly state whose results have agreed with the observations here.

L227-230: I am not sure where I can find the evidence showing the decrease and effect. Please describe the location of this information specifically.

L241-242: Both Figure 3 and Table S1 do not show OC/EC ratios. Please provide those.

L246-247: There would be many possible reasons for this variation, and I do not agree with the statement of "The elevated wet-season. . .". I recommend either to remove this sentence or provide the evidence to justify this possibility.

L255: I recommend to replace "a pulse" with a different term. How about occasional input?

L271: Avoid symbols of arrow and infinity in the text.

L277-278: In my opinion, descriptions of "d13C enrichment" and "13C/12C ratio enrichment" are not correct, but enrichment in (or with) 13C is the correct description. Please consider this throughout the text.

L275-281: I am not sure the point the authors want to make in this paragraph. Occurrence of chemical reactions in the particle? Please re-write it to make the point clearer.

L284: Please provide the degree of "shift" specifically. Also replace "around" with "$\sim$" sign, or vise versa.

L286: Not sure what agreed in. Please make this specific.

L287* Please define "ITCZ".

L303: State Figure 3 when discussion the results shown somewhere.

L330: Replace "between" with from.

L331-334: As I mentioned earlier, I recommend to analyze possible variations in more detail, then provide this fingerprinting d13C that gives feasible results. Choice of this value without other possibilities will lead a biased interpretation.

L340-342: I am not sure why the authors can say so. Please provide the evidence (observations or references demonstrating such characterizations).

L355-356: Does Figure 6 show 71% of carbonaceous aerosols from savanna fire in the dry season? I do not know where I can find this information in the figure.

L360-390: I recommend to rewrite "Outlook" because some indirect topics, such as CO2 source and brightening stuff, are referred with many references.

Table 1: Correct “BC/EC”.

Figure 1: As I mentioned earlier, provide two plots for two seasons.

Figure 2: The scales of y-axes are overlapped. Please fix those.

Figure 4c: It is interesting to see such a relationship. If extrapolating the curve to zero of TC, which is originated from plant burning, what the intercept value would be? I recommend to briefly discuss this value in the text.

Figure 5: I recommend to refer the references for d13C of C3, C4, and fossil in the footnote.

Figure 6: Add the horizontal line of zero for panel A. By the way, all figure captions should be under the figures. Are those just the style of ACPD?

Table S1: Refer inorganic components in the table heading as well.

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