

Our responses to the Reviewer are organized as: Reviewer comment in italic and response in blue regular font. The changes made in the manuscript refer to the new version (Page, Line), in bold.

This paper presents a two-year monthly time series of measurements of aerosol chemistry (organic and inorganic) from a sampling site in the Ugandan mountains, and area sensitively placed to record seasonal variations in aerosol sources in sub-Saharan Africa. The measurements include radiocarbon and stable carbon isotope determinations on a subset of samples from across the wet and dry seasonal cycle. These measurements enable the delineation of major changes in aerosol source related mostly to a changing input of aerosols derived from savanna burning. While there is nothing that is particularly novel in the results compared to the range of previous work in the region over the last decades, the results do contribute to a growing and useful body of aerosol data from this very large region of the world. The study has been well conducted and a series of robust results generated that clearly show the seasonal impact of savanna burning on local aerosol composition. The source apportionment using carbon isotopes is particularly valuable. I do not have any substantive issues with the analysis or interpretation.

We thank Reviewer 1 for the overall supportive comments and constructive feedback. We have updated the manuscript accordingly (see details below) and believe that the manuscript is significantly improved.

In response to Reviewers 3 and 4 we have made one larger change in the paper: we have implemented an expanded Bayesian MCMC technique, in which the isotope correlations with TC are used to constrain the sources. This method is based on our work from Martens et al. (2019) – see elaborations in response to Reviewer 4. The method is described in the updated Section 2.5 and the results are discussed in the updated Section 3.5. In connection to this, we have also discussed three sensitivity scenarios, w.r.t, C₄ and fossil $\delta^{13}\text{C}$ endmembers.

New/changed figures and tables:

Figure 1: We have updated Figure 1, now with back-trajectory arrival heights at 100 m.a.g.l., and 500 m.a.g.l. as a new Figure S1. In the submitted version the arrival heights were (by mistake) 10 m.a.g.l, and the latitude was slightly offset. We think 100 and 500 are more representative, while they also in good agreement.

Figure 4: We moved the $\Delta^{14}\text{C}$ vs TC plot to a new **Figure 5**, in which we also added a $\delta^{13}\text{C}$ vs TC plot.

The previous **Figure 5** (2D isotope plot) is the new **Figure 6**.

We have updated the previous **Figure 6** with the results from the new MCMC approach, and this is the new **Figure 7**.

New Figure S1: back trajectories at arrival height 500 m.a.g.l.

New Figure S2: $\Delta^{14}\text{C}$ vs TC and $\delta^{13}\text{C}$ vs TC from the new Bayesian MCMC source apportionment method,

New Figure S3: A sensitivity analysis of the new Bayesian MCMC source apportionment strategy w.r.t. number of data points.

New Figures S4-S6: computed fractional source contributions from 3 alternative endmember scenarios; sensitivity tests.

New Table S2 with updated fractional source contributions from the new MCMC approach.

New Tables, S3-S5: results from the MCMC-based source apportionment from the 3 alternative endmember scenarios.

Reviewer comment: The paper is possibly somewhat long for the amount of data it presents, and does make some slightly overblown claims about the importance of the results in the context of regional environmental sustainability, climate and health that could be reduced in scope.

We agree, that the implications may have been over-stated, while we still maintain that the health- and climate impact serves as an important motivation for the study, even though these are not directly addressed. We have removed related parts in the Abstract, the introduction and in the Outlook.

We have updated the manuscript accordingly, including the specific comments by reviewer and the other reviewers, as well as the specific points raised in the annotated supplement to these comments, see below.

I have also identified a large number of grammatical issues on the attached pdf Please also note the supplement to this comment: <https://www.atmos-chem-phys-discuss.net/acp-2019-1027/acp-2019-1027-RC1-supplement.pdf> Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/a>

Responses to specific comments

We here respond to specific questions or more substantial changes.

Page 1, line 22.

‘major uncertainties of the, e.g., climate and health impacts’

We change this to:

‘In this paper we use ground-based observations to address the currently large uncertainties in source-resolved emission estimation of carbonaceous aerosols.’

Page 2, Lines 21-23.

Page 5, line 117.

Yes, 1M HCl.

Page 6, line 130.

‘The accuracy of the measurement ranges from 7% for 1 mg·L⁻¹ of carbon solution to 3% for concentrations higher than 2 mg·L⁻¹ of carbon.’

We have clarified this as:

‘The accuracy of the measurement ranges from 7% (70 µg L⁻¹) for 1 mg L⁻¹ of carbon solution to 3% for concentrations higher than 2 mg L⁻¹ of carbon (corresponding to 60 µg L⁻¹).’

Page 6, Lines 138-140

Page 8, lines 190-197.

‘The meteorology of Rwanda is governed by the East African monsoon, with peak rainfalls in in April and November. There are thus two dry seasons, December-January-February (DJF) and the main dry season June-July-August (JJA). The dry periods in SSA are characterized by extensive biomass burning. During DJF the fires mainly occur to the north of Rwanda, and during JJA to the south (Fig. 1). Savannas are the main biomes in SSA, covering ~ 65% of the landmass, and are the main source of fire emissions (Cahoon et al., 1992). Located in a highly elevated region,

Rwanda is, broadly speaking, surrounded by savanna regions, except to the west, where the tropical rainforests of Africa are located.'

We moved this section to the M&M section 2.1 describing the station.

Page 5, Lines 96 to 103.

Page 9, line 214.

We have changed 'aerosol regime' to 'aerosol sources and atmospheric processing'

Page 10, Line 241-242

Page 10, line 242.

The OC/EC-ratio is unitless.

Page 11, line 271.

TC is defined in the limit $0 \leq TC < \infty$, and is not expressed in units of percentage.

References

Martens, J., Wild, B., Pearce, C., Tesi, T., Andersson, A., Bröder, L., O'Regan, M., Jakonsson, M., Sköld, M., Gemery, L., Cronin, T.M., Semiletov, I., Dudarev, O.V., Gustafsson, Ö.: (2019) Remobilization of Old Permafrost Carbon to Chukchi Sea Sediments During the End of the Last Deglaciation. *Glob. Biogeochem. Cyc.* 33, 2-14, doi: [10.1029/2018GB005969](https://doi.org/10.1029/2018GB005969).