

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.

It's a nice little study that adds to the much-needed set of measurement data in SSA. Novel in the sense data in SSA are missing, but otherwise a mainly normal study. Ambient aerosols have been collected at a remote site in Rwanda and analyzed for their content of carbonaceous species and some ions. The regional influence of savannah fires is nicely confirmed by the isotope measurements. Apart from a few clarifications I would like the authors to do in the manuscript, the analysis and discussion part of the manuscript is OKI, but the claims of impacts on climate and health are outside the focus of the paper. Just as reviewer #1 points out, there are some grammatical mistakes that needs the C1 attention of the authors. I will not repeat the things #1 already mention but add to the list below.

We thank Reviewer 2 for the overall supportive and constructive comments and suggestions. We have incorporated the majority of the suggestions (see below) and think the paper is now 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.

Line 22: While possible fixing the comment from #1, please see to that the final sentence is possible to understand. The current is not.

This sentence now reads.

‘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 21-22.

L 33: Remove “, an SSA background site”

Done, thanks.

L 57-58: Could this be expanded a bit? What is and why is it distinct?

We have replaced this sentence with

‘Ground- and airborne chemical characterization from this and other campaigns suggest a rather distinct aerosol chemical composition, including elevated BC, K^+ and NO_3^- concentrations (Table 1).’

PAGE 3, LINE 54-56.

L 68. I miss a climate related reference for the claim. If climate should still be part of the paper.

We believe that climate is an important motivator for investigating source dynamics of aerosols in SSA.

We added the reference IPCC, 2013.

Page 4, Line 66

L 71-72: I miss a reference for the claim.

We added the reference to Liousse et al., 2015.

Page 4, Line 70.

L 99 + 102: If only nighttime measurements were done, how can this help a strategy to study diurnal variation? And is that a strategy of this study?

Diurnal variability was not an aim of this study. We collected 7 days samples, but the sampler was only on during the night.

We now write:

‘Night-time only (1AM to 6AM) was conducted to minimize the effects of local emissions and day-time local atmospheric chemistry and to increase likelihood to capture the regional, free troposphere, signals. This strategy is supported by high temporal resolution investigations of the diurnal cycle of, e.g., BC (DeWitt et al., 2019). Each sample was collected over a period of 7 days.’

Page 5 Lines 107-111

L 150: Location of the institute?

We added location as:

“National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility at the Woods Hole Oceanographic Institute (Falmouth, Massachusetts, USA)”

Page 7. Line 158

L 155-158: If the intention is to show the reader how F_{bio} is calculated, why not rearrange Eq. 1 into $F_{bio} = \dots$?

Yes, the Equation now reads:

$$f_{bio} = \frac{\Delta^{14}C_{sample} - \Delta^{14}C_{fossil}}{\Delta^{14}C_{bio} - \Delta^{14}C_{fossil}}$$

L 167: I think a “=” before +57 would make “ $C_{bio} + 57 \pm 52 \text{‰}$ ” more readable.

We agree. We have updated accordingly.

Page 7 Line 175

Section 2.5: The heading and the text are presenting the content in different order. Why?

We agree this is inconsistent and switched the order.

Page 9 Line 215

Throughout the manuscript: Don't mix "season" and "period". Stick to "season". Change "emissions sources" to "emission sources". "Emissions" appears in other combinations as well, where I think the plural "s" should be removed.

We agree and have updated accordingly throughout the manuscript.

L 205: BT not defined. Could be written in full.

We agree and now write 'back-trajectories' throughout the paper.

L 220: I normally avoid "as well as other factors" as it is not very helpful for the reader. I think most readers of a aerosol paper understands that we are not in full control of everything in ambient measurements and thus this addition is not needed.

We agree and removed this part.

L 222-223: remove one set of "has also". I also miss a reference for this claim.

Removed. We added Gao et al. (2003) and Formenti et al. (2003), from Table 1, as references.

Page 10, Lines 250-251.

L 237: What activity? Volcanic? Human?

Yes, it was unclear and now reads:

'but with no clear linkage to an increase in volcanic SO₂ emissions.'

Page 11, Line 264.

L 242: Fig. 3 shows EC/TC, not OC/EC ratio.

Yes, we choose to plot EC/TC for parallel construction in Figure 3. However, OC/EC is more commonly discussed. We have changed the text as:

‘Here, the EC/TC shows a distinct seasonality (Fig. 3 and Table S1). More commonly analyzed, though, is the OC/EC ratio ($=(\text{TC}-\text{EC})/\text{EC}$), with elevated levels during the wet season (11 ± 3) compared to the dry season (7 ± 3 ; Table S1).’

Page 11 Lines 268-270

L 247: Have you considered that there is more plants material that can contribute to the OC in wet season? Can windblown dust during dry season increase the EC concentrations and thus the TC?

We removed the sentence “The elevated wet-season OC/EC may indicate increased relative influence of local SOA formation.” As we think it is too speculative.

L 248: Is SO₂ really elevated during wet season? It looks more random. Volcanic influence?

The SO₄²⁻/TC levels are more scattered than some of the other ratios, but overall it is higher during the wet seasons (0.29) compared to the dry season (0.22). It is not unlikely that this reflects regional volcanic activity.

We added:

“... including potential volcanic input of SO₂”

Page 11 Line 278

L 265: Occasionally? Within your analysis errors they are all above +20.

We agree and removed ‘occasionally’.

L 277: 12C?

Good point. We now write:

‘An overall enrichment in ¹³C has been found in aged air masses in South Asia, especially for WSOC’

Page 12 Lines 310-311

L 295-298: To me the numbering of the points looks strange. (1.) etc. I would prefer it written in text instead.

We removed the numbering and now write:

‘First, the $\delta^{13}\text{C}$ -value is not an exclusive source marker, but is also affected by atmospheric processing (e.g., photo-chemical oxidation and secondary formation). Second, the main source categories must be defined and distinguishable using carbon isotopes. Third, the source-values of the isotope-signatures, the endmembers, and their natural variability need to be established.’

Page 13 Line 327-331

L 356-357: Reference to Supplement Table S1? If it is the results in this study that is referred to.

We agree and added reference to Table S1.

L 360: Remove one “that”

Thanks. Done!

L 365: Why is the site suddenly called a mountain background site? Why not use the name again?

We agree, and replaced with RCO.

L 389: “expected rapid change”. Is that something that is already happening? If so, can references from 1990’s be used for comparisons in the paper? Like “g” in Table 1.

We agree, in principle: The rapid change is seen in many sectors, including emission inventory estimates (Lioussé, 2015). However, given the few ground-based data it is hard to evaluate such trend from observational data; especially when measured at different sites, and campaign-wise.

Figure 1, and text. The blue lines in the figure comes from the Arabic peninsula. Can you still claim you have no influence of flaring from that region?

We have updated the back-trajectories with what we think are more realistic arrival heights, 100 and 500 m.a.g.l, compared to 10 m.a.g.l. This suggests a slower influence from the Arabic peninsula.

New Figures 1 and S1.

We now write:

‘However, given the distances to the RCO station and the prevailing wind directions, emissions from flaring are not expected to affect the site, while the $\delta^{13}\text{C}$ -signatures for gas-flaring are strongly depleted in ^{13}C ($\delta^{13}\text{C} < -38\text{‰}$; Winiger et al., 2017) and even a small contribution would shift the observed values significantly.’

Page 14 Lines 252-355.

Figures 2, 3, 5, 6: “The November 2014 to April 2015 gap is due to a lightning strike” only makes a reader curious. I suggest reformulating it. Like “Instruments were hit by lightning resulting in a data gap November 2014 to April 2015”

Thanks. We have updated accordingly.

Figure 3: In the bottom graph there is an extra “+” after TC in the legend.

Thank you. We have removed the +.

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)

Winiger, P., Andersson, A., Eckhardt, S., Stohl, A., Semiletov, I.P., Dudarev, O.V., Charkin, A., Shakova, N., Klimont, Z., Heyes, C., Gustafsson, Ö.: (2017) Siberian Arctic black carbon sources constrained by model and observation. *Proc. Nat. Acad. Sci.* doi: [10.1073/pnas.1613401114](https://doi.org/10.1073/pnas.1613401114).