

Response to Editor Decision

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General comment :

The authors have engaged with the reviewer comments well. The study is very interesting and in a data poor area. I don't have issues with the study itself. However, the authors have not yet adequately engaged with the research in the introduction to provide a summary of what is known. There is even a place where the paper has found the opposite of what is stated. This needs to be updated and improved, mostly in the introduction, but a few places outside of it. In addition, in the discussion, one point that needs clarity is which of these sources are possible sources over a full year, and which are possible sources during this short sampling campaign? I understand why they included the discussion of more sources, and it is helpful to better understand the full system. However, I recommend they are clearer in their writing throughout that section that these are sources not near where the back trajectories went, but could be sources. This is a point they do get to, but I think for readers who are not familiar with this area, it could be confusing. I have noted where I thought some additional information to clarify this point would help.

I have a few other comments below that I recommend are considered and addressed.

We wish to thank the editor for a thorough review of the manuscript and the amount of new relevant literature specific to our study region. For the value added to our manuscript, a special acknowledgement was made for the insightful suggestion at the end of the manuscript.

Line 43 : I recommend including SAFARI literature that discusses burning in southern Africa.

References to the SAFARI campaigns were added line 44 of the revised manuscript.

And

I recommend being more precise. In South Africa, the rain in the summer rainfall region follows this dry season, but not all of South Africa. Some discussion of this and relevant citations are here: Roffe, S. J., Fitchett, J. M., & Curtis, C. J. (2019). Classifying and mapping rainfall seasonality in South Africa: a review. *South African Geographical Journal*, 101(2), 158–174. <https://doi-org.uplib.idm.oclc.org/10.1080/03736245.2019.1573151>

The provided reference is not open access and/or cannot be accessed by authors. However, this manuscript broadly focuses on the south-eastern region of Africa and not only on South Africa specific emission regions; hence, we believe that a detailed description of the variabilities in meteorological provinces is not relevant to this manuscript and would add complexity to the introduction with no further use in the discussion section. We have addressed one comment on specific weather events at the time of the study, lines 355-358 of the revised manuscript, where this information was deemed useful to justify our results.

Line 53 : I recommend adding in the SAFARI references that first described this.

References to the SAFARI campaigns were added line 54 of the revised manuscript.

Line 56 : This wording confuses me some as I understand aeolian transport to be associated with dust transport. How is it linked to smoke? Is it that they are occurring at the same time? I recommend this is just clarified in this sentence to avoid confusion by readers.

The term aeolian transport was changed to “atmospheric transport”, lines 57-58 of the revised manuscript, so as to include emissions from any atmospheric source including but not limited to dust. “More locally, atmospheric transport from Madagascar has also been demonstrated across the Mozambique Channel during the dry season (Kumar et al., 2014).” In order to respond to the editor question’s, emissions of smoke (from fire emissions) were indeed shown to contain a large percentage (up to 60%) of dust in previous literature (see Hamilton, D.S. et al. 2022. *Earth, Wind, Fire, and Pollution: Aerosol Nutrient Sources and Impacts on Ocean Biogeochemistry. Annual Review Marine Science*. 14:303-330. <https://doi.org/10.1146/annurev-marine-031921-013612>); however, this question is out of the scope of the current manuscript.

Lines 60-62: This is not the correct reference for this statement. The emissions of coal fired power stations are quantified in many papers and reports. I would recommend rather referencing papers that estimate emissions and using more precise wording than "massive". The direct PM from coal fired power station (if this is what it is referring to) are not the largest emissions of concern. Also, I believe this statement is about South African more than all the southern African countries.

The sentence was modified as well as the reference, lines 60-64 of the revised manuscript. “In addition, South Africa is the 7th largest coal-producing and coal-consuming country in the world. The production (mining) and use of coal, be it for industries, energy production or for domestic burning, results in emissions of airborne hazardous volatile particles (such as lead, Pb, cadmium, Cd, or mercury, Hg) threatening both the environment and human health (Wang, 2023; Zerizghi et al., 2022a).” Indeed here, we are pointing to coal use and production-related emissions in general. Such emissions are pointed out as they include volatile trace elements which are used as tracers in our study.

Line 62 : “to the east coast” was changed to “located to the east of South Africa” line 64 of the revised manuscript

Line 64 quote 1: This is a quote from the paper "The decreasing trends in PM emissions (Figure 2) reflect the improving removal efficiency of the PM abatement technology (electrostatic precipitators and flue gas conditioning plants at Komati and 3 units at Grootvlei initially, and fabric filter plants at Medupi, Camden and 3 units at Grootvlei) as defects are rectified and performance is optimised. The extremely low PM emissions at Grootvlei from 2017 are due to the fabric filter plant retrofits on units 2, 3 and 4. "This study does not support this statement. Thank you for reporting inconsistency in our wording. Indeed, here we do not wish to point out particulate emissions (“PM10”) but rather the emissions of selected volatile trace elements of interest. The sentence, lines 64-66 of the revised manuscript, has been reworded to fit the reference’s point. “For example, the activity of thermal power plants located to the east of South Africa, showed a four to five folds rise over the last decade owing to the increased demand of power generation (Morosele and Langerman, 2020; Zerizghi et al., 2022).”

Line 64 quote 2: This article estimates SO₂, Pb, and Hg emissions, not PM. Thus, it also doesn't support this statement.

The statement was revised to specifically target those volatile trace elements we are using as tracers (especially Pb and Cd). See revised sentence in the response to quote 1 above.

Line 154 : Annual dust deposition values are described in the results. How were these samples scaled up to yearly deposition should be included here.

The deposition velocity constant used includes a temporal component ($V_d = 1.2 \text{ cm s}^{-1}$). This constant term is commonly used in atmospheric trace element deposition studies to assess annual deposition of particles assuming a constant deposition rate year-round. Here, the deposition velocity per second is also expressed in m/d (1036.8 m d^{-1}) and extrapolated to the whole year by multiplying the flux by 365. The sentence and equation were revised lines 155-160 of the revised manuscript “Based on assumptions made in previous studies (Baker et al., 2016; Marsay et al., 2022), a constant deposition velocity (V_d) of 1.2 cm s^{-1} (or $V_d = 1036.8 \text{ m d}^{-1}$) was used to calculate F_{Dust} following Eq. (1):

$$FDust = \frac{T_{Al} \times V_d}{[Al]_{UCC}} \times 365 \quad (1)$$

where T_{Al} is the total Al concentration measured in aerosols, and $[Al]_{UCC} = 8.04\%_{w/w}$ (McLennan, 2001). The resulting $FDust$ flux was expressed in $\text{mg m}^{-2} \text{ yr}^{-1}$ by multiplying the daily flux by 365 days.”

Line 316: It is not clear to me which years these modelled values are for. Including this would help to support the statement on line 323 about little inter-annual variability.

A line was added to Table 2 to display the year of the simulation when clearly stated in the reference. In 2 references, the exact years were not clearly stated and were referred as “present day” (relative to the publication date) or simply mentioned as “unknown”

Line 319 : April can still have a lot of rain in the summer rainfall region of South Africa. Was April indeed, dry this year during measurements? It could help to include some indication of this here.

A sentence was added on particular weather events that occur on the year 2022. However, these events are highly specific to 2 locations in our study area and may not be significant in terms of flux variability compared to when accounting for dust/fire emissions. Lines 355-358 of the revised manuscript

“In addition, high precipitations associated with both the flood events in the Kwa-Zulu Natal region of South Africa in mid-April 2022 (Radio France, 2022) and the Jasmine tropical storm on the western coast of Madagascar in late April 2022 (Meteo France, 2022), could result in reduced dust entrainment in the atmosphere, in turn contributing to lower fluxes calculated during our study period.”

Line 320 (references): Archibald paper should be included as this has timing of fires as those listed here re about dust

Archibald, S., Scholes, R. J., Roy, D. P., Roberts, G., and Boschetti, L.: Southern African fire regimes as revealed by remote sensing, *Int. J. Wildland Fire*, 19, 861–878, <https://doi.org/10.1071/WF10008>, 2010.

The reference was added to the reference list and to the revised manuscript line 343.

Line 387 : I would recommend deleting "so-called" as this can have the connotation that it is not the correct word for these. But coalfields is correct. However, they are not close to any of the back trajectories shown to have occurred. I would clarify then how could they be a source to these aerosols?

The word « so-called » was deleted line 400 of the revised manuscript.

A justification was added to address the back-trajectory analysis issue raised here lines 416 – 420 of the revised manuscript “Air-mass trajectory analysis shown in Figure 2 show existing trajectories, in green (group II samples) and in red (group III samples), passing near such coalfield regions. While the coalfield located further north to Lesotho on the South African country do not appear as clear source in the air-mass trajectory analysis (Figure 2), these trajectories only represent clustered samples trajectories and do not represent the exact trajectories for air-masses included in each individual aerosol sample collected” (rather they indicate a common broad direction for the cluster).

Line 392: This is not true. It is uncertain, but not unknown. There is literature working to estimate emissions, often at smaller scale than national. However, there are global emission inventories, such as DICE Africa that does estimate the emissions as well as DACCIWA.

DICE: Marais and Widiomyer 2016, [pubs-acsc.org/uplib-idm.oclc.org/doi/10.1021/acs.est.6b02602](https://pubs.acs.org/uplib-idm.oclc.org/doi/10.1021/acs.est.6b02602)

DACCIWA Keita, S., Lioussé, C., Assamoi, E.-M., Doumbia, T., N'Datchoh, E. T., Gnamien, S., Elguindi, N., Granier, C., and Yoboué, V.: African anthropogenic emissions inventory for gases and particles from 1990 to 2015, *Earth Syst. Sci. Data*, 13, 3691–3705, <https://doi.org/10.5194/essd-13-3691-2021>.

The term unknown was replaced by “uncertain” line 423 of the revised manuscript and reference was added for indication. In the absence of quantification for the specific components we investigate in this study, we do not display number for the associated emissions.

Line 402: This reference seems to be missing from the list. I wanted to look if this is a key source during the time sampled or in general.

I find it a little difficult in this section to track what in general could be a source of dust to this region and what would be from these samples that hysplit showed had minimal continental contribution. I recommend making it clearer here in this section.

The reference was added to the reference list

Line 472: Same comment as above that these are only dust and not fire timing papers. I recommend adding the Archibald paper.

The reference was added line 507 of the revised manuscript

In the conclusion, a sentence was also added on the temporal variability of the identified sources.

“While the atmospheric sources pointed out in our study are likely to run over the full year, time-series analysis of the identified chemical tracers’ atmospheric loading is necessary to confirm such hypothesis.”

In addition, Table 1 of the main manuscript and Table S4 of the supplementary material were adapted according to Dr Katja Gänger’s comment on the use of colours in our manuscript.