Dust deposition fluxes at the gateway to the Southern Ocean: investigating the use of lithogenic tracer measurements in aerosols collected in Tasmania, Australia

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Response to reviewers

Reviewer 1: Zongbo Shi

This is a solid paper. It proposed a multi-tracer method to estimate dust fluxes. It then estimated the dust flux at a strategic location in the Southern Ocean. The methodology is robust. The results are well presented and the conclusion is well justified.

I only have a few minor comments for consideration.

1. Title – be more concise. Words like "investigating" are a waste of space

Good suggestion, thank you. In addition, a second Reviewer suggested to refine the geographical position of the study location in the title. Amended title to: "On the use of lithogenic tracer measurements in aerosols to constrain dust deposition to the ocean southeast of Australia ".

2. Line 12-12: I suspected that you mean the flux estimated is between a peak dust deposition event and a low event. If so, the writing as it is does not represent this. Please clarify this and revise accordingly.

Changed to: "Lithogenic flux estimates showed annual dust deposition maxima during austral summer, following the Australian dust storm season, and annual minimum deposition flux over winter." lines 11-13 of the revised manuscript

3. Line 16-19: This is a bit wordy. The first part of the sentence appears to repeat the previous sentence. The main point appears to be something like: the data provided here will help to constrain model estimates of ….

Changed to: "The data provided here will help to constrain model estimates of southern hemisphere atmospheric deposition fluxes and their subsequent impact on global ocean biogeochemical cycles." lines 13-15 of the revised manuscript

4. Line 136: explain why 125 samples only for 6 years? E.g., give information on the sample duration and frequency.

The sentence at lines 136-141 has been removed and inserted above after the sentence at line 113-116, which explains sample duration/frequency. Additional information was also added at lines 118-120 of the revised manuscript regarding the number of samples included in this study. "Samples suspected for contamination or that were significantly wet at the time of recovery were discarded and sampling was suspended in the winter time of 2017, 2018 and 2019."

5. Line 196-197: how an aliquot be DRY sieved?

Changed to: 'Ten milligrams of each soil sample was dry sieved…" line 211 of the revised manuscript

6. I am sorry if I have missed but how the total mass of aerosols was estimated? This is important to mention as it determines the accuracy of the Fe/total aerosol mass ratio. It should be noted that there may well be sea salt and other natural/anthropogenic aerosols. This could reduce the total Fe content in the total aerosol. Similar applies to Al. This may partially explain the low mean Al/Fe contents in aerosols. It would be great if a mass closure (e.g., sulfate, nitrate, sea salt, OC, EC, dust etc.) is given if such data are available. This comment is also relevant for points raised in the paragraph starting line 286.

In the Methods section, line 168 onwards, the deposition flux calculation is detailed. As mentioned lines 186-189, we used the "Similar to other studies reported in the literature, a single-tracer dust (lithogenic) deposition flux estimate, $F_{Lith(X)}$, was calculated by dividing F(X) by the average abundance ($[X]_{UCC}$, wt%) of the element X in the UCC as reported in McLennan (2001); Al = 8.04%, Fe = 3.5%, Th = $1.07x10^{-3}$%, Ti = 0.41% following equation (2)". That way, the total aerosol mass was estimated using the relative abundance of each tracer in the upper UCC as per McLennan (2001).Unfortunately no additional measurements other than trace metals were available for all samples of the time series dataset. That way, no direct calculation on the total aerosol mass was possible in this study.

Also added lines 169-171 of the revised manuscript: "Additional measurements on the collected aerosols (e.g.; carbon and major ion analysis) were not available for this study, so intrinsic calculation of the total aerosol mass on each individual aerosol filter using these parameters was not possible."

7. Table S2 – total Fe content in soil appears to be very low. Yes, there may be spatial variabilities. But could there also be a possibility of the size dependence? The size cut here is about 63 um. And in reality, you are unlikely going to see many particles of that size at the sampling location due to long range transport (not to say that it is impossible). I suggest that the authors look at literature and see how other studies have estimated the total Fe content, both in terms of the size cut of the particles, and methodology. Secondly, can you show all other elements you measure for all soil samples. They are very useful reference data for future research.

Methodology aimed at representing particle grain size capable of being uplifted from the source region and transported as aerosol towards our aerosol sampling site at Mt Wellington. Our methodology matches other studies undertaken in Australia as Strzelec et al 2020 (see lines 211 - 212) as well as the National Geological Survey Australia size cut for soil fine fraction (<75um, de Caritat et al., 2009).

A Table S3 was added to the supplementary documents, including measurements of other trace metals in the selected NGSA soil samples. This data will not be discussed in the main manuscript as it is out of the scope of our study. Mention to the new table S3 was added lines 243-245 of the revised manuscript “Metal concentrations in individual NGSA soil samples analysed in this study are reported in the supplementary Table S2 (lithogenic tracers) and in Table S3 (other analysed trace metals not discussed in this study).”

8. Line 266: this is an interesting point. Later sentences supported this argument. Are there representative back trajectories that you can show to support this point? It would be good to have the back trajectories from high and low dust flux seasons.

HYSPLIT air-mass trajectory frequency (AMBT) analysis was added to the supplementary documents Figure S1, in place of the initial Figure S1. Using the HYSPLIT AMBT model shows no significant seasonal difference in the observed wind influences at our sampling station. This may be due to the coarse resolution of the model which emphasises the prevailing westerly winds originating from the Southern Ocean. However, contrasting information is shown using mean wind data from Australian Bureau of Meteorology (BoM) for our sampling station (shown in a new supplementary Figure S2). Figure S2 seems to show increased winds originating from the North (from the Australian mainland) in the months of January through to March.

As both observations from HYSPLIT and from BoM diverge, further discussion was added lines 283-289 of the revised manuscript :

“While enhanced air-masses originating from the Australian mainland cannot be observed in the summertime using HYSPLIT model (supplementary Figure S1), the Australian Bureau of Meteorology reports increasing southwards blowing winds at our sampling station from January through to March (supplementary Figure S2). Such discrepancies emphasise the complex regional wind pattern influencing our sampling station and highlight the need to consider other parameters such as seasonal changes in environmental conditions at the source region when investigating aerosol entrainment and transport. ”

In addition, literature supporting the south eastwards transport of dust from Australian mainland is referred to in the manuscript :

Mackie D.S. Biogeochemistry of iron in Australian dust: From eolian uplift to marine uptake. *Geochem. Geophys. Geosyst.* **2008**, *9*, Q03–Q08. doi.org/10.1029/2007GC001813

Baddock M., K. Parsons, C. Strong, J. Leys, G. Mctainsh, Drivers of Australian dust: a case study of frontal winds and dust dynamics in the lower lake Eyre basin. Earth Surf. Process. Landforms, 40 (2015), pp. 1982-1988, 10.1002/esp.3773

Che Y., B. Yu, and K. Bracco, Temporal and spatial variations in dust activity in Australia based on remote sensing and reanalysis data sets, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2023-1710, 2023.

Tang W., J. Llort, J. Weis *et al.* Widespread phytoplankton blooms triggered by 2019–2020 Australian wildfires. *Nature* 597, 370–375 (2021). https://doi.org/10.1038/s41586-021-03805-8

9. Line 316 – spelling error

Changed ‘ound’ to ‘found’ in the revised manuscript

10. Figure 3 – please mention briefly what ratios are being used? UCC or Australian soil results?

This is already stated in the final sentence of the caption.

11. In Figure 1, would it be appropriate to consider add the locations by Strzelec et al. (and any other studies) where the dust flux was estimated?

In the revised manuscript, Figure 1 now displays the study location of previously reported dust deposition fluxes.

12. There are mentions of fire and related dust. This is an interesting point but I do wonder whether you can provide any supporting evidence, such as higher K+ concentrations. I presume you haven’t analysed levoglocosan?

Levoglucosan or potassium analysis are beyond the scope of this study. Due to the lack of evidence of the proposed hypothesis, we decided to remove the suggested impact of fire emissions and only rely on NGSA database supporting higher Fe/Al ratios in Tasmanian soil. Amended sentence line 296-300 of the revised manuscript “Summertime Fe/Al ratios in kunanyi/Mt Wellington aerosols were slightly higher (Fe/Al =0.72) than the Australian soil measurements. This can be explained by increased contribution of local soil emission from Tasmania under drier weather conditions as the NGSA database shows higher Fe/Al ratio (on average 0.7, n=21 samples) in Tasmanian soil compared to other soil across Australia (Caritat and Cooper, 2011; Caritat, 2022).”

13. Paragraph starting 397: I wonder whether you can compare the estimated fluxes with more modelling studies. I think there are several global modelling studies of dust deposition fluxes. For example, Mahowald et al. 2005. https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2004GB002402

The Mahowald et al. (2005) reference was not included in the manuscript as it was judge to be a contemporary literature to Jickells et al. (2005) and reporting similar dust fluxes (1.4- 2.7 $mg/m^{-2}/d^{-1}$). The dust flux reported in Mahowald et al. (2005) falling within the 1.4 – 5 $mg/m^{-2}/d^{-1}$ range now discussed in the revised manuscript line 370, the suggested literature was added to the reference list

Mahowald, N. M., Baker, A. R., Bergametti, G., Brooks, N., Duce, R. A., Jickells, T. D., et al. (2005). Atmospheric global dust cycle and iron inputs to the ocean. *Global Biogeochemical Cycles*, **19**(4), GB4025. https://doi.org/10.1029/2004GB002402.

14. Uncertainties: I agree that the multi-tracer method is more reasonable and better than a single tracer estimate. However there are still uncertainties and I suggest that you add a paragraph or section to discuss specifically about all the possible uncertainties, e.g., ratios, deposition velocities. It does not affect the conclusion of this paper but it will help readers to understand the manuscript better, and to use it more appropriately. If you can give an estimate of the uncertainty range, e.g., 2 times, that would be helpful. But I suspect it is not going to be an easy job. It may be worth mentioning that models still have large uncertainties so the uncertainties from observation-based flux estimates are still relatively small.

Uncertainty associated with the use of a set deposition velocity in our flux calculation was already mentioned in the manuscript Materials and Methods section although a quantitative uncertainty is now provided lines 180-184 of the revised manuscript "It should be mentioned that a factor of 3 uncertainty was previously attributed to the use of a set deposition velocity as it does not account for specific particle size in different aerosol samples or for specific atmospheric conditions such as humidity and wind speed at the collection time (Baker et al., 2016; Winton et al., 2016; Duce et al, 1991)."

Uncertainty associated with the use of the lithogenic tracer's abundance in the UCC (as a mean to calculate total aerosol mass) is also mentioned in the paragraph starting line 483 onwards of the revised manuscript. A paragraph summarizing uncertainty surrounding our dust flux calculation was added to the conclusion of the revised manuscript, lines 507-514. "Dust deposition fluxes calculated in this study hold some uncertainties of a factor 3 and a factor 2 due to the use of a set deposition velocity and the assumption of metal abundance as per the average UCC, respectively …"

15. Conclusions – this is rather long. Most of the points in the conclusions have already been mentioned in abstract. I wonder whether the final section as "Atmospheric implications" might be more useful to readers. Can you tell us a bit more about the implications of the results reported here? You mentioned nutrient inputs – are the inputs, in different seasons, likely to be important for ocean plankton and biological pump?

As an abstract is a condensed version of a complete paper, the authors feel it is reasonable to have some repeated conclusions in the Abstract and Conclusions sections. However, we have re-written the conclusion to make it shorter and more readable. Implications of the study are further developed in the final paragraph.

16. Can you also have a short paragraph, perhaps at the end of the "atmospheric implications" if you decide to have one, about what future research should be

done? You did mention somewhere in the text about the research needs – but they could be at one place of the manuscript.

Future research needs, including but not restricted to additional time-series data and winter samples, are now mentioned in the last paragraph of the conclusion section.