Dear Editor,

On behalf of all co-authors, I thank you for considering this paper for submission to ACP. We have taken the reviewers' comments into account as far as possible.

Specific comments from the reviewers are addressed below.

Please note that the author list has changed, with two additional co-authors; the author contribution section and the acknowledgement have also been updated accordingly. The data associated with this study have been deposited in the Easy Data repository (www.easydata.earth), and we will provide a DOI as soon as it becomes available.

Reviewer 1

General Response: We thank the reviewer for the constructive comments. We have addressed these comments point by point below.

Comment R1.1: The spatial representativeness of source categories, such as Free State, Skukuza, Hope Mine, and Kalahari BOT, is less clear. The authors should provide, either in the main text or the Supplement, the number of samples for each source type, their spatial coverage, the criteria used for site selection, and evidence that these sites correspond to active or potential dust-emission hotspots.

We have updated **Lines 152-174** to reflect the reviewer's comments as follows:

"A total of 40 soils were selected for experiments (Fig. 1 and Table S1) to represent major natural dust-source areas across southern Africa, as reported in the literature (Ginoux et al., 2012; Prospero et al., 2002; Vickery et al., 2013) and subject to sample availability. A particular focus is on the Namib Desert, with its extensive ephemeral riverbed system, which is a recognised dust source area (Ginoux et al., 2012; Vickery et al., 2013; von Holdt et al., 2017). To characterise sources across the Namib Desert, we used two soil samples from the Central Namib Gravel Plain and the Kuiseb riverbed, collected at the Gobabeb Namib Research Centre. In addition, we collected soil samples from the terminal reaches of the Huab, Kuiseb, and Omaruru riverbeds along 1-km transects at 100-m intervals, resulting in 12, 8, and 13 points, respectively. A sample from the Etosha Pan was included in our study as another significant dust source (Buch and Rose, 1996; Vickery and Eckardt, 2021), though it is more variable and intermittent than the Namib Desert (Bryant, 2003; Bryant et al., 2007; Haustein et al., 2015). Dust sources in the Kalahari Desert tend to emit less than those in the Namib Desert. We included one sample from the southwestern Kalahari in Botswana, which is a significant emerging source (Bhattachan et al., 2012; Bhattachan et al., 2013; Vickery et al., 2013), as well as two samples from areas of the Kalahari Desert north and south of the Okavango Delta in northwestern Botswana, which is an important but more local source (Bullard, 2004; Vickery et al., 2013).

This study also provides the first characterisation of anthropogenic dust sources (four source-soil samples in total, Fig. 1 and Table S1) that are emerging in southern Africa as a result of anthropogenic pressure and climate change (Eckardt et al., 2020; Ginoux et al., 2012; Vickery et al., 2013; Vos et al., 2025). These include an agricultural soil in the Free State, identified as a key area of agricultural dust emissions (Eckardt et al., 2020; Trisos et al., 2022; Vickery et al., 2013). In addition, two savannah soils were collected at Skukuza in the Kruger National Park, South Africa, from an area periodically subjected to prescribed fires, representing bare soils that can generate dust emissions after vegetation is removed by fire, a condition frequent in northeastern and eastern South Africa (Strydom and Savage, 2016). Finally, we used one sample from an abandoned copper mine, Hope Mine in Namibia, to explore the composition of mineral dust from mining activities, which are widespread across southern Africa and have also been identified as emerging sources of mineral dust (Vos et al., 2025)."

Please note that the details of the laboratory-generated aerosol samples and soils are provided in Table S1 of the supporting information as stated in **Lines 145-147**:

"Details of the laboratory-generated aerosol samples from this study are provided in Table S1, including the locations and geographic coordinates of the corresponding soils, the system used for the aerosol generation, and the availability of chemical and mineralogical data."

Comment R1.2: The study uses GAMEL and CESAM to generate dust aerosols, but their possible effects on elemental or mineralogical fractionation are not fully discussed. The authors should clarify whether the two systems may introduce systematic differences in the generated dust composition. If common samples were analysed using both systems, a direct comparison would be useful.

We have updated **Lines 269-279** to reflect the reviewer's comments as follows:

The methods developed for the GAMEL and CESAM systems to generate PM_{10} dust aerosols are based on the same apparatus and the same physical principle, which consists of simulating wind-driven soil particle movement by shaking soil within a glass flask, thereby causing the breakage of soil grains and agglomerates to release fine dust particles (see reference papers by Di Biagio et al., 2014; Di Biagio et al., 2017; Lafon et al., 2014). Furthermore, to exclude method-induced differences in the elemental and mineralogical composition of the resulting dust aerosols, we compared the elemental compositions of dust aerosols generated by CESAM and GAMEL from a similar soil type, using the Kuiseb riverbed samples. Figure S2 compares the chemical composition of Kuiseb dust generated by CESAM, using different samples collected from a riverbed transect in the terminal reach, with that of Kuiseb dust generated by GAMEL, using a riverbed sample collected at the Gobabeb Namib Research Centre. The elemental concentrations were normalised to Al to account for variations in the mass generated. The two sets can be considered equivalent within the experimental standard deviation, represented by the 10% uncertainty of XRF measurements.

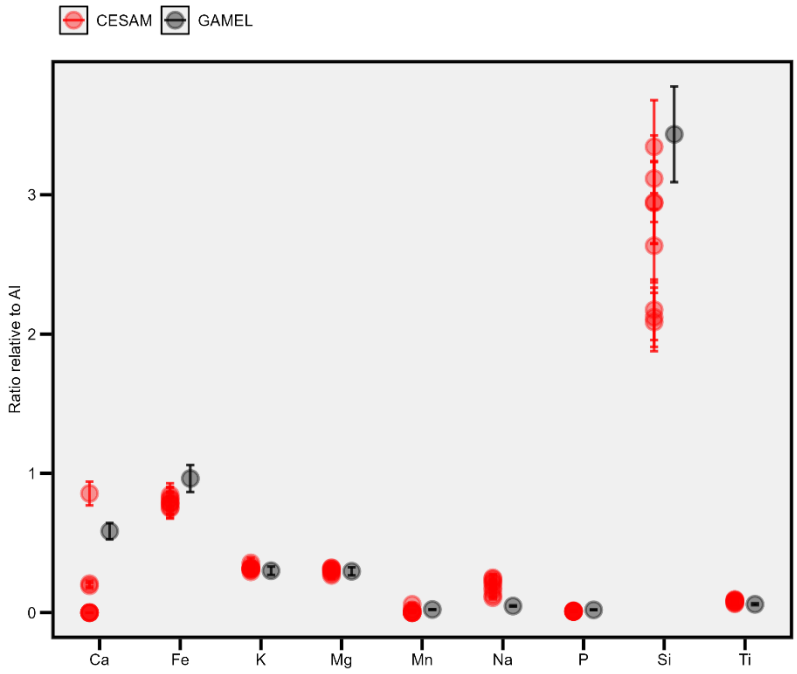

Figure S2: Elemental compositions of dust aerosols generated by CESAM and GAMEL from a similar soil type, using the Kuiseb riverbed sample. In red, the chemical composition of Kuiseb dust generated by CESAM, using different samples collected from a riverbed transect in the terminal reach, while in black, that of Kuiseb dust generated by GAMEL, using a riverbed sample collected at the Gobabeb Namib Research Centre. The elemental concentrations were normalised to Al to account for variations in the mass generated. The error bars represent a 10% uncertainty in XRF measurements.

Comment R1.3: The total aerosol mass was estimated from the sum of major elemental oxides. This approach may introduce biases for samples enriched in carbonates, salts, sulfates, hydrated minerals, or water. The authors should compare the oxide-sum mass with gravimetric filter mass, if available, or discuss the potential bias caused by excluding components.

We acknowledge that estimating dust aerosol mass by summing major elemental oxides can introduce uncertainty, as this approach may not fully capture the true dust composition compared with the gravimetric method. However, it remains a widely used approach for retrieving dust aerosol mass from elemental composition. To assess the differences between the two approaches, our previous study (Caponi et al., 2017) compared reconstructed mass with gravimetric mass measurements of laboratory-generated dust aerosols from global sources, spanning a wide range of mineralogical compositions. The reconstructed mass agreed well with the gravimetric mass, with an average difference of less than 10%.

Comment R1.4: The manuscript suggests that carbonate-rich Southern African dust may promote heterogeneous reactions, particle ageing, hygroscopicity, and CCN activity. This is plausible, but the current evidence is mainly mineralogical. The authors should either provide quantitative or semi-quantitative support, or relevant literature-based constraints.

The potential role of Southern African dust (SAf) dust in promoting ageing and CCN activity has been further discussed and supported by the literature, as follows:

Lines 613-623: Compared with documented major dust sources in the Southern Hemisphere and Northern Africa, SAf dust is enriched in calcium carbonate minerals, which play a significant role in atmospheric heterogeneous reactions, promoting the uptake of gaseous pollutants and the ageing of dust (Krueger et al., 2004; Laskin et al., 2005; Sullivan et al., 2009; Tang et al., 2015). During atmospheric transport, calcite and dolomite particles in SAf dust can interact with urban and marine aerosols, undergoing heterogeneous reactions with reactive trace gases such as HNO_3, SO_2, HCl, and VOCs. This process can produce atmospheric ageing products, including highly hygroscopic species such as $Ca(NO_3)_2$, $CaCl_2$, and $Ca(HCOO)_2$, which can act as efficient CCNs, whereas mineral dust has very low hygroscopicity when emitted (Krueger et al., 2004; Laskin et al., 2005; Sullivan et al., 2009; Tang et al., 2015). However, further research is needed to assess the hygroscopicity and CCN activity of SAf dust and their evolution during atmospheric transport, which is particularly important because aged SAf dust aerosols with high hygroscopicity and CCN activity may influence the formation of stratocumulus clouds over the northern Bay of Benguela.

Comment R1.5: The Si/Al, (Ca+Mg)/Al, and K/Al ratios are reasonable tracers for distinguishing arid western coastal sources from more humid inland eastern sources. However, Fig. 5 shows considerable overlap among some source categories. The authors should consider applying multivariate or classification methods, such as PCA, hierarchical clustering, or random forest classification, to evaluate how well these ratios discriminate between source regions.

Following the reviewer's suggestion, we conducted a Principal Component Analysis (PCA) of our integrated dataset, which included the elemental ratios (Ca+Mg)/Al, K/Al, Si/Al, and Fe/Al measured in this study, as well as data from the literature. Observations from salt pans with distinctive features were excluded because they dominated the PCA and reduced variation among other samples. PCA was conducted using the prcomp() function from core CRAN R software.

The results are shown in Fig. S7. The first two principal components account for around 75% of the variance. Principal Component 1 (PC1) was predominantly associated with the ratios K/Al and (Ca+Mg)/Al, which are nearly orthogonal to Si/Al and Fe/Al, the latter primarily associated with Principal Component 2 (PC2).

Source regions occupy distinct positions in PCA space. The Namib GP and Kalahari NAM clusters intersect and are distributed across quadrants I and IV, near the (Mg+Ca)/Al and K/Al vectors,

suggesting these samples have higher (Mg+Ca)/Al and K/Al ratios and may be associated with weakly weathered material rich in carbonate and primary aluminosilicate minerals, as represented by PC1. The Kalahari BOT sample, agricultural dust from the Free State, and savannah samples from Skukuza, grouped as Eastern source areas, cluster in quadrant II, near the Si/Al and Fe/Al vectors, indicating higher Si/Al and Fe/Al ratios, which could be linked to more weathered source soils rich in silica and Fe oxide, as represented by PC2. Hope Mine shows characteristics similar to those of the Eastern source cluster. The riverbed samples are concentrated in a narrow group within quadrant III, showing minimal variation along PC1 but a wider distribution along PC2 in the opposite direction to the Si/Al and Fe/Al vectors. This indicates lower Si/Al and Fe/Al ratios, suggesting weaker weathering compared to Eastern source areas.

Overall, these results further confirm that Si/Al, (Ca+Mg)/Al, and K/Al ratios can be used as mineral dust source tracers in southern Africa and can separate arid coastal sources from more humid, inland eastern source areas. Note that all anthropogenic sources are predominantly associated with PC2, which we attribute to more weathered source soil. Most of the source areas, except for Hope Mine, are located in more humid regions that have experienced greater weathering in the past and current climates. However, anthropogenic activities are concurrent and can also contribute to the processing of source soil and enhance weathering.

The discussion above was included in the supporting information as **Text S3**.

Please note that **Lines 525-528** in the main text have been updated as follows:

"These results are further corroborated by a Principal Component Analysis (PCA) of key elemental ratios, conducted using the prcomp() function from core CRAN R software on our integrated dataset. This dataset included the elemental ratios (Ca+Mg)/Al, K/Al, Si/Al, and Fe/Al measured in this study, as well as data from the literature. The PCA results are discussed in Text S3."

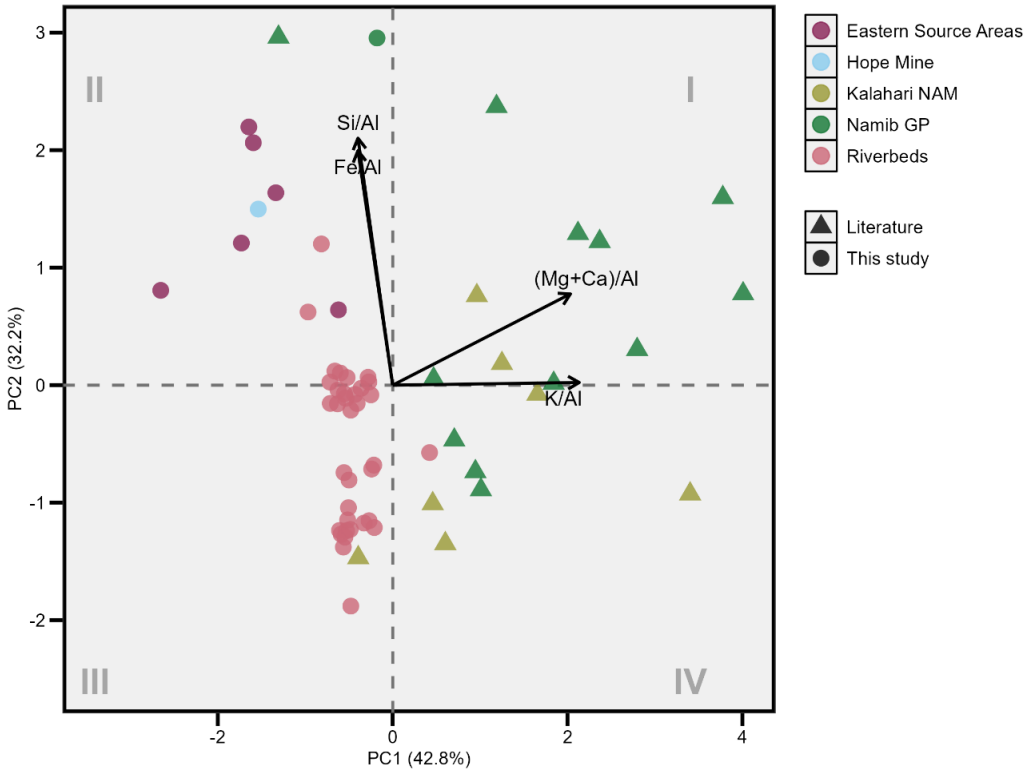

Figure S7. Principal Component Analysis (PCA) of key elemental ratios observed in Southern African dust aerosols. Namib GP data include this study, Di Biagio et al. (2017), and Qu (2016). Kalahari NAM data are from Qu (2016). The Kalahari BOT sample, agricultural dust from the Free State, and savannah samples from Skukuza were grouped as Eastern source areas. The PCA results are discussed in Text S3.

Reviewer 2

General Response: We thank the reviewer for the insightful comments. We have addressed these comments point by point below.

Comment R2.1: I do wonder if the paper would fit better in a journal such as Global Biogeochemical Cycles. I think ACP will be just fine but the authors should consider whether they will lose their geochemistry audience by publishing in an atmospheric journal.

We thank the reviewer for this suggestion and agree that our manuscript is of great interest to the geochemistry community. We intend to submit it to Atmospheric Chemistry and Physics (ACP), specifically to the Special Issue on Southern African aerosols, to reach this audience effectively. Furthermore, our previous ACP publications on related topics, including studies on Icelandic dust (Baldo et al., 2020) and coal fly ash (Baldo et al., 2022), have been highly cited across both the atmospheric science and geochemistry communities. We consider continuing the research into impacts, and in that respect we will seek to target the biogeochemistry audience more appropriately.

Comment R2.2: The authors can also comment on the dust emitting potential of the different sources using work from (Prospero et al., 2002)

Lines 152-174 have been updated to reflect the reviewer's comments. Please see *Response R1.1*.

Comment R2.3: Methods: I'm unclear about how the dust was generated and how comparable the two methods are. Please clarify the comparability of the two methods to a reader unfamiliar with either method. I'm especially unclear how the CESAM works. A brief description needs to be added. For each system: what gas is used to suspend dust in the GAMEL and how much flow is used? Since more flow is used to collect the filters than is stated to generate the dust, what kind of dilution or makeup flow is used in both systems?

The methods developed for the GAMEL and CESAM systems to generate PM_{10} dust aerosols are based on the same apparatus and the same physical principle, which consists of simulating wind-driven soil particle movement by shaking soil within a glass flask, thereby causing the breakage of soil grains and agglomerates to release fine dust particles (see reference papers by Di Biagio et al., 2014; Di Biagio et al., 2017; Lafon et al., 2014). Furthermore, to exclude method-induced differences in the elemental and mineralogical composition of the resulting dust aerosols, we compared the elemental compositions of dust aerosols generated by CESAM and GAMEL from a similar soil type, using the Kuiseb riverbed samples. Figure S2 compares the chemical composition of Kuiseb dust generated by CESAM, using different samples collected from a riverbed transect in the terminal reach, with that of Kuiseb dust generated by GAMEL, using a riverbed sample collected at the Gobabeb Namib Research Centre. The elemental concentrations were normalised to Al to account for variations in the mass generated. The two sets can be considered equivalent within the experimental standard deviation, represented by the 10% uncertainty of XRF measurements. Please also refer to *Response R1.2*.

In addition, a brief description of both the GAMEL and CESAM systems has been added to **Section 2.2**:

"The "Générateur d'Aérosol Minéral En Laboratoire" (GAMEL, Lafon et al., 2014) was used to generate dust aerosols from parent soil samples. The GAMEL apparatus consists of three main components: a generation system, a PM_{10} cyclone for size selection of particles with aerodynamic diameters less than 10 μm, and a collection system for dust aerosols on filters connected to a pump via tubing.

For the soil samples collected at the terminal parts of the Huab, Kuiseb, and Omaruru riverbeds, dust aerosols were generated using the Multiphase Atmospheric Experimental Simulation Chamber

(CESAM, Wang et al., 2011). This facility has been widely used to characterise the chemical and physical properties of natural mineral dust aerosols (Baldo et al., 2023; Baldo et al., 2020; Caponi et al., 2017; Di Biagio et al., 2014; Di Biagio et al., 2019; Di Biagio et al., 2017). An external dust-generation system is attached to the 4.2 m^3 multi-instrumented environmental stainless-steel reactor. Stainless-steel tubes connect the chamber body to custom-made samplers, which are linked to a pump that extracts dust aerosol from the chamber onto filters."

The details on carrier gases and dilution flow for both the GAMEL and CESAM systems have been added in **Lines 240-253**:

"The dust aerosol generator in GAMEL consists of a mechanical wrist-action shaker (Agitest®, 8 mm amplitude) that simulates the wind-driven movement of soil grains, thereby reproducing the effect of sandblasting. Around 1-3 g of 1000 µm-sieved soil was agitated in a Büchner flask for 3-12 min at shaking frequencies of 500-650 cycles/min, depending on the observed dust generation efficiency of the soils. The system uses ambient air, filtered through an HEPA filter connected to the generation flask. The airborne dust aerosols released from the parent soils were drawn through tubing into a size-selective cyclone device (URG-2000-30EA, URG Corporation, Chapel Hill, NC, USA) connected to a vacuum pump operating at a controlled total flow rate of 30 L min^{-1} for PM10 sampling.

The dust aerosol generator coupled to CESAM comprised a sieve shaker (Retsch AS200) that agitated a Büchner flask containing about 10 g of soil (sieved to 1000 µm and dried at 100°C for about 1 hour). The dust particles released by mechanical shaking were injected into the simulation chamber via a nitrogen carrier gas (10 L min^{-1}) for about 20 minutes. The chamber was filled with synthetic air prior to each experiment, while a fan at the bottom of the chamber ensured homogeneous mixing of the particles (Wang et al., 2011). The particles remained suspended for approximately 3 hours, during which time they were characterised using several online and offline instruments. After dust aerosol injection, Synthetic air is also used to compensate for dilution caused by air-sampling instruments."

Comment R2.4: Methods: Were filter blanks also taken and similarly analysed?

Both the GAMEL and CESAM systems were manually cleaned before experiments on different soils. To ensure a particle-free environment, dust concentration was verified before each experiment using online OPC and SMPS measurements. Blank filter samples were also collected and analysed to check for possible contamination.

Please note that this paragraph was added at **Lines 290-292**.

Comment R2.5: How reliably was Al detected by your XRD methods? This is key to state, as Al is the primary element that most data is ratioed to.

We agree that quantifying elemental Al is key to the paper's results. Elemental Al was measured by X-ray fluorescence (XRF) and not X-ray diffraction (XRD). X-ray fluorescence (XRF) is commonly used to analyse Al and other crustal elements in atmospheric aerosols, providing a direct, non-destructive method for analysing particles on filters (Bilo et al., 2024). The uncertainties and quantification limits for elemental mass concentrations measured by X-ray fluorescence are reported in Section 2.2.3. The uncertainty of the XRF measurement is 10%, with quantification limits of 1-4 µg for Ca, Fe, K, and Nd, and 0-0.5 µg for the other elements (including Al). In addition, the measurements were corrected for self-attenuation effects affecting light elements, including Al. Therefore, we believe that normalising elemental concentrations by Al concentration is sufficiently robust.

Comment R2.6: Figure 5: I cannot distinguish the different studies. Can similar colors be used for the same region but different marker shapes be used to show the spread in other studies vs the current study?

Figure 5 was updated to reflect the reviewer's comments.

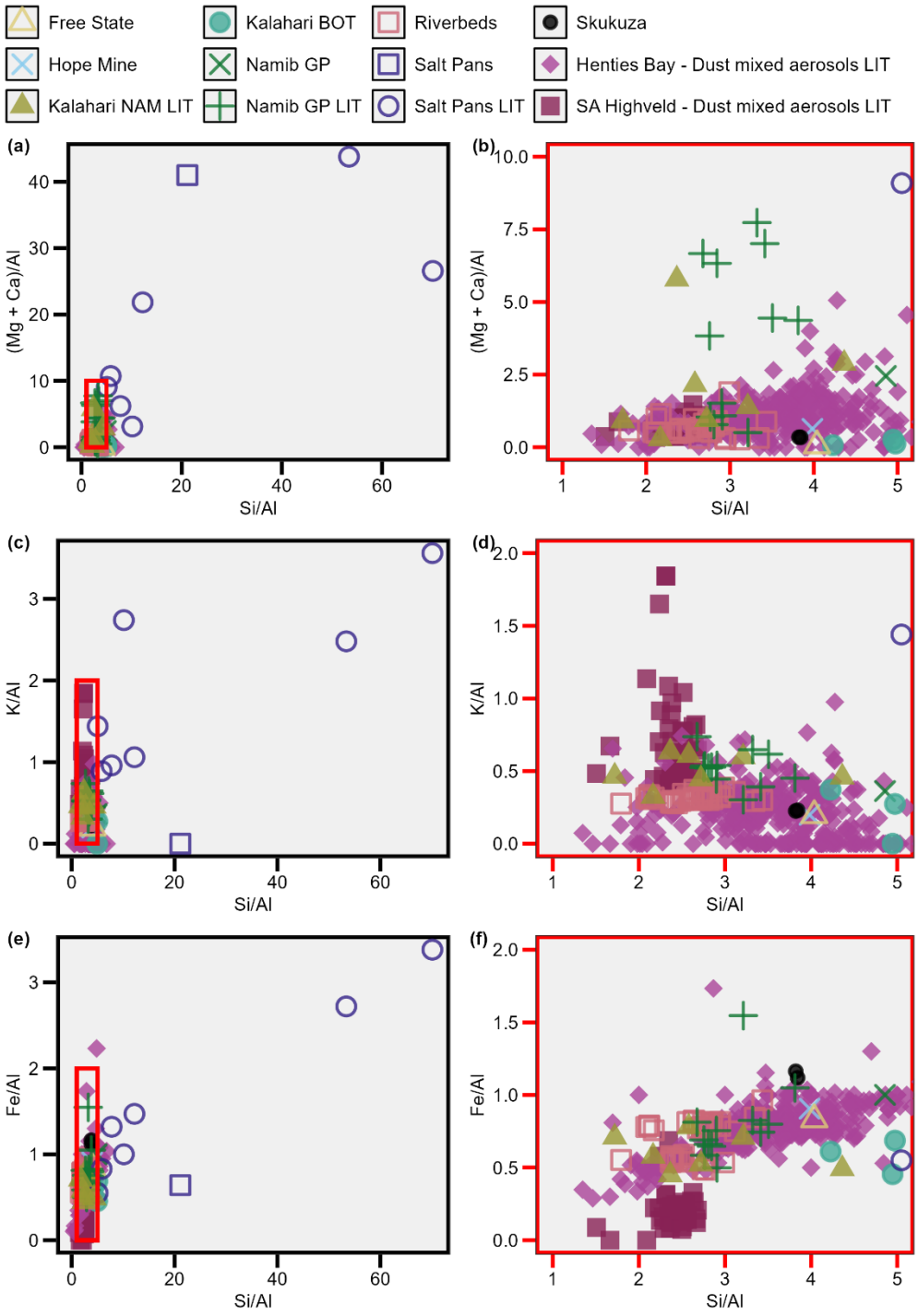

Figure 1. Mineral dust tracer variability across Southern Africa. The red rectangle indicates the zoomed area in the right-hand panels. For the reader's sake, the error bars are not shown. Namib GP data include this study, Di Biagio et al. (2017), and Qu (2016). Kalahari NAM data are from Qu (2016). Salt Pans data include this study and Engelbrecht et al. (2016). Ground-based observations of dust mixed aerosols over the Namibian coast (Desboeufs et al., 2024; Formenti et al., 2025; Klopper et al., 2020) and the South African Highveld (Baldo et al., 2025) are included for comparison. Data source references are summarised in Table S3. LIT indicates data from the literature.

Comment R2.7: Line 518-520: is the low detection of Na simply due to the methodology?

We agree with the reviewer that global dust sources are depleted in Na, partly because techniques such as PIXE and XRF, which were used to assess the elemental composition in the selected studies, have low sensitivity to Na, as the fluorescence yield rapidly decreases as the atomic number decreases (Beckhoff et al., 2007).

Comment R2.8: Heterogeneous reactions on carbonates (calcite and dolomite) are mentioned and the carbonates are cited as CCN. The authors should amend their discussion of carbonates to reflect that while sodium containing carbonates are very hygroscopic, calcite and dolomite are not so their ability to act as CCN is limited to only those that have undergone chemical aging (Gaston et al., 2017; Laskin et al., 2005; Sullivan et al., 2009).

We agree with the reviewer that dolomite and calcite minerals may act as CCN after chemical ageing. **Lines 613-623** have been updated to clarify this aspect (*see Response 1.4*)

Comment R2.9: The authors also mention the bioavailability of micro and macro nutrients. Typically dusts require some chemical aging to make these nutrients available. Carbonates act to buffer the acid processing needed to solubilize P and Fe for example (Nenes et al., 2011; Stockdale et al., 2016) so the authors should clarify this important point.

We agree with the reviewer that carbonates buffer the acid processing needed to solubilise nutrients in mineral dust. Nonetheless, an increased Fe solubility of 20% has been observed in dust aerosols over the Namibian coast, attributed to atmospheric processing by marine aerosols. Although SAf dust is enriched in carbonates, its role as a source of dissolved nutrients varies with source, transport, and ageing processes. Further research is essential to understand how these ageing processes influence nutrient dissolution in SAf dust.

Lines 624-630 have been updated to reflect the reviewer's comment:

Additionally, aged SAf particles can contain dissolved essential micronutrients (Fe, P, and Mn), which can affect biogeochemical cycles in marine ecosystems as also suggested by recent research (Belelie et al., 2025; Gittings et al., 2024). An increased Fe solubility of 20% has been observed in dust aerosols over the Namibian coast, attributed to atmospheric processing by marine aerosols (Desboeufs et al., 2024). Although SAf dust is enriched in carbonates that can buffer the acid processing required to solubilise nutrients in mineral dust (Ito and Feng, 2010), its role as a source of dissolved nutrients varies with source, transport, and ageing processes. Further research is essential to understand how these ageing processes influence nutrient dissolution in SAf dust.

Comment R2.10: The authors could expand their discussion of the anthropogenic dusts particularly in the discussion and abstract. The high Cu, P, and Fe is very interesting and has a lot of implications for marine biogeochemical cycles, particularly if, as the authors suggest, these sources are increasing in spatial extent.

We appreciate the reviewer's comment. A short paragraph was added to the discussion, and a short sentence was added to the abstract to complement our conclusions:

Abstract, Lines 35-36: In particular, emerging anthropogenic dust can be distinguished by its high content of certain nutrients.

Lines 515-519: This study analysed only a limited number of mineral dust samples from anthropogenic sources, so it may not reflect the variability across different anthropogenic dust source types. However, our results show distinct features, such as high Cu and P in mining dust and high Fe in savannah and agricultural dust, which may have important implications for marine biogeochemical cycles in the

present and especially in the long term, as these sources are emerging due to climate change and anthropogenic pressure.

Comment R2.11: MINOR POINTS

Figure 1 has a typo "mentionned" should be fixed

Line 560: typo "Cal" should be "Ca"

All minor comments were addressed.

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