

# Global Biogeochemical Cycles<sup>\*</sup>

## RESEARCH ARTICLE

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### Special Collection:

EMIT: The Earth Surface Mineral Dust Science Investigation

### Key Points:

- Earth Surface Mineral Dust Source Investigation mineralogical distributions are used for the dust source elemental distribution
- Non-dust sources are included for natural and anthropogenic sources
- Observations of Al, Ca, Fe, Si, and Ti are compiled for land-based stations and cruise data and compared to modeled values

### Supporting Information:

Supporting Information may be found in the online version of this article.

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## Global Model Estimates of Atmospheric Al, Ca, Fe, Si, and Ti From Dust and Non-Dust Aerosols Informed by EMIT Surface Mineralogy and Evaluated Against Observations

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**Abstract** Atmospheric deposition of micro-nutrients like Fe has been shown to be important for ocean biogeochemistry. The largest source of atmospheric Fe and other elements (e.g., Ca, Al, Si, and Ti) is desert dust, although there are significant non-dust sources in some regions (e.g., combustion, sea salts, volcanoes). However, past estimates of these elements have been substantially uncertain due to limited information about the composition of the desert source regions. Here we use elemental distributions estimated from new Earth Surface Mineral Dust Source Investigation (EMIT) observations, which provide mineralogical composition at the surface of the Earth based on imaging spectroscopy measurements from the International Space Station. We focus on total elemental amounts, not on the soluble fraction. We add in other sources of these elements (anthropogenic and natural) and compare to a compilation of available surface concentration data from stations over land and from shipborne observations. The combined observational and model synthesis provides new information about the distribution and deposition of these elements. Our results suggest that the modeled distribution is similar to available observations, but discrepancies still exist in both natural desert dust regions as well as regions dominated by anthropogenic sources. Comparisons between the model estimated Ca/Al ratios and observations in some dust dominated regions suggest an underestimate of Ca/Al ratios. Global budgets for Ca, Al, Fe, Si, and Ti suggest that desert dust remains the dominant source, although volcanic and anthropogenic contributions are important in some regions. Changes in elemental distributions since preindustrial times were also estimated.

**Plain Language Summary** We provide estimates of the atmospheric concentrations and deposition of aluminum, calcium, iron, silicon, and titanium using models and compiled observations from land and ship-based studies. These estimates provide improved information about the distribution and allow future studies to consider the possible impacts of atmospheric transport of these elements.

## 1. Introduction

Atmospheric deposition of biogeochemically relevant elements on land and ocean ecosystems can provide important nutrients and enhance growth or be toxic and inhibit ecosystem functions (Chapin et al., 1986; Likens et al., 1996; Paytan et al., 2009; Rodríguez et al., 2023; Schlesinger, 1997). Human emissions of fossil fuel derived particles, as well as the anthropogenic modifications to aerosols such as from wildfire or arid regions (Hamilton et al., 2018; Kok et al., 2023; Mahowald et al., 2018; Van Marle et al., 2017), have substantially altered atmospheric aerosol inputs to land and ocean ecosystems, potentially modifying biogeochemistry and carbon uptake in these systems (Duce et al., 2008; Jickells & Moore, 2015; Jickells et al., 2005; Mahowald, 2011; Mahowald et al., 2011). Most of the earlier studies on atmospheric deposition and their impact have focused on nitrogen, iron, or phosphorus (e.g., Chien et al., 2016; Duce et al., 2008; Jickells et al., 2005; Mahowald et al., 2008; Vet et al., 2014), although other elements are also likely to be important for ocean and land biogeochemistry (e.g., Baker et al., 2006; Jickells & Moore, 2015; Lu et al., 2024; Mackey et al., 2012; Mahowald et al., 2017, 2018).

Desert dust is the most abundant component of atmospheric aerosols by mass and provides many important elements to land and ocean ecosystems (Mahowald et al., 2017, 2018). The major elements in desert dust are aluminum (Al), calcium (Ca), iron (Fe), silicon (Si), and titanium (Ti), each combined with oxygen and hydrogen in the mineral form, although the quantity of these varies slightly in different arid or desert regions depending on the mineralogy in the source regions (Desboeufs et al., 2005; Formenti et al., 2008; Rodríguez et al., 2020; Zhang et al., 2015).

Atmospheric Fe deposition to the open ocean is likely important both for phytoplankton growth in Fe-limited regions (Boyd et al., 2007, 2017; Martin, 1990) as well as the higher Fe requirements for nitrogen-fixing organisms (Capone et al., 1997; C. M. Moore et al., 2013; K. Moore et al., 2006; Tagliabue et al., 2017), and because

of this, atmospheric Fe deposition to the oceans has been extensively studied (Duce et al., 1991; Hamilton et al., 2021; Ito et al., 2019; Mahowald et al., 2009; Matsui et al., 2018; Myriokefalitakis et al., 2018; Opazo et al., 2025).

Besides Fe, the other dominant elements in dust (Al, Ca, Si, and Ti) may also play an important biogeochemical role once they are deposited. Al is one of the dominant elements in dust composition comprising about 7%–8% by mass (Zhang et al., 2015). In the ocean, Al plays a potentially important role in diatom opal remineralization, linking it to Si availability for diatoms (Mahowald et al., 2018; Orians & Bruland, 1986; van Hulst et al., 2014). Ca plays important roles in ocean biogeochemistry as a main sink for the carbonate ion, as well as a nutrient for ocean biota although atmospheric deposition from land to ocean is likely smaller than riverine inputs (Fantle & Tipper, 2014). Si is important for ocean biogeochemistry, as many important ocean organisms (e.g., diatoms) require it in substantial amounts to form shells and other structural materials (Tréguer et al., 2021), and atmospheric deposition is estimated to represent about 10% of the new inputs from riverine sources (Schlesinger, 1997; Tréguer et al., 2021). Ti in the ocean is thought to have limited biotic interactions (Orians et al., 1990) and has been used to help interpret paleorecords (e.g., Murray et al., 1993). On land, both Ca and Al play important roles in buffering soil pH (Fantle & Tipper, 2014; Schlesinger, 1997), although in most soils atmospheric deposition is unlikely to be important compared to local bedrock or soil sources (Fantle & Tipper, 2014; Schlesinger, 1997). Ca in the atmosphere can play an important buffering role, modifying the aerosol pH (Baker et al., 2021).

The abundance of each element varies in different soils, creating differences in atmospheric dust composition which can be used as a geochemical tracer of different dust sources (Grousset & Biscaye, 2005). These variations in composition also change the biogeochemical, radiative, and chemical properties of the dust that are important to consider in modeling studies (Hand et al., 2004; Journet et al., 2008; Okin et al., 2004; Scanza et al., 2015). Previously, soil mineralogy and elemental composition estimates used Food and Agriculture Organization (FAO) soil type maps, combined with approximately 1,000 soil samples (Claquin et al., 1999; Journet et al., 2014). Alternatively, studies have used elemental soil concentrations to create maps of dust source composition (Zhang et al., 2015). This approach extrapolates a few data points, mostly from non-arid agricultural regions, to millions of grid cells thousands of kilometers away in semi-arid and arid regions, which then causes substantial uncertainties in the resulting mineral and elemental composition estimates (Li et al., 2021; Okin et al., 2004; Scanza et al., 2015; Wong et al., 2021). One of the novel contributions of this research is that we take advantage of new mineralogy maps provided from the Earth Surface Mineral Dust Source Investigation (EMIT) (Green et al., 2023). This space-borne mission has measured surface mineralogy in arid and semi-arid regions across the globe at 60 m pixels which are combined and aggregated into  $0.5^\circ \times 0.5^\circ$  grid boxes (Okin et al., 2023).

The atmospheric deposition of non-ferrous elements, when considered, has previously only focused on dust deposition, ignoring other non-dust sources of these elements (e.g., Measures & Vink, 2000; van Hulst et al., 2014; Zhang et al., 2015). Globally-averaged estimates suggest important anthropogenic and natural sources (e.g., combustion, sea salts, volcanoes), which may differ at the regional level (Mahowald et al., 2018; Nriagu, 1989; Nriagu & Pacyna, 1988; Pacyna & Pacyna, 2001; Schlesinger, 1997). While in some contexts the soluble fraction is more important, here we focus on the total elemental amounts, since there are much more data and knowledge of these fractions (Mahowald et al., 2018).

In this paper we simulated the atmospheric emissions, transport, and deposition of total (not only soluble) Al, Ca, Fe, Si, and Ti, the dominant elements in dust, using the new EMIT-derived soil mineralogy data set, but also including sources from anthropogenic and other natural sources. From this comprehensive approach, we can better understand both where and when the non-dust sources are important, as well as how these elements are likely to have changed since preindustrial times. In Section 2, we introduce the models and data used in the paper. Section 3 compares our results against available observations for the distribution of the elements. We then show global budgets and estimate the change in deposition of these elements since preindustrial times to highlight the land and ocean ecosystems where the impact of human perturbations to these elements could be the largest on their biogeochemistry. Finally, we present our summary and conclusions.

## 2. Methods

### 2.1. Atmospheric Modeling

Elemental aerosol concentration simulations were conducted using the appropriate parameterizations within the Community Atmosphere Model, version 6 (CAM6) and the atmospheric component of the Community Earth System Model (CESM2) coordinated at the National Center for Atmospheric Research (NCAR) (Hurrell et al., 2013; Liu et al., 2011; Scanza et al., 2015). The model calculates three-dimensional transport by nudging toward MERRA2 winds (Gelaro et al., 2017) and wet and dry deposition for gases and aerosols. Simulations were conducted at approximately  $1^\circ \times 1^\circ$  horizontal resolution with 56 vertical layers for 4 years, with the last three years (2013–2015) used for the analysis (Computational and Information Systems Laboratory, 2019). These simulations are very similar to those used for previous aerosol studies (e.g., Lu et al., 2024; Mahowald, Li, Vira, et al., 2025).

The default model includes prognostic dust, sea salts, black carbon, organic carbon, and sulfate aerosols using a modal aerosol scheme (Li et al., 2021; Liu et al., 2011, 2016). In order to improve the simulation of coarse mode particles, the coarse model size parameters were returned to the values in the CAM5, and there was an improvement to the dry deposition scheme (Li et al., 2022). Fossil fuel emissions of sulfate, organic carbon and black carbon follow the Climate Model Intercomparison Project 6 emission scenarios for present day for 2010 (Gidden et al., 2019). All aerosols are subject to dry and wet deposition (Liu et al., 2011; 2016).

### 2.2. Desert Dust Elemental Distributions at the Sources

Desert dust or mineral aerosols are entrained into the atmosphere in dry, sparsely vegetated regions subject to strong winds. The Dust Entrainment and Deposition scheme (Zender et al., 2003) is used with the updated Brittle Fragmentation Theory (Kok et al., 2014) and improved incorporation of aspherical particles for optics and deposition (Huang et al., 2021; Kok et al., 2017; Li et al., 2022). Eight different minerals were simulated for dust, and the elemental contribution from each mineral, as estimated in Menut et al. (2020), is used to estimate the Al, Ca, Fe, Si, and Ti elemental emissions and distributions (Table S1 in Supporting Information S1). The global annual dust emission in our simulation is approximately  $2,900 \text{ Tg yr}^{-1}$ , with a global mean dust optical depth in the visible band ( $0.44\text{--}0.63 \mu\text{m}$ ) of  $\sim 0.03$ .

This model version is identical to the one used in Li et al., 2022, except for the changes in the EMIT soil mask, described below, and the model performs for the dust, very similar to previous versions in terms of comparisons to aerosol optical depth, surface dust concentrations and deposition (Figure S1 in Supporting Information S1). Note that it is often difficult for dust models to simulate all three: optical depth, concentrations and deposition accurately (Huneeus et al., 2011), and here we emphasize the dust tuning that accurately describes the deposition since we are interested in biogeochemistry impacts of the deposition.

NASA's EMIT mission provides estimates of soil mass fraction at  $0.5^\circ \times 0.5^\circ$  spatial resolution for the minerals chlorite, calcite, dolomite, goethite, gypsum, hematite, illite/muscovite, kaolinite, montmorillonite, and vermiculite (Brodrick et al., 2025; Green et al., 2020). EMIT uses the spectral radiance measured from the International Space Station's orbit (Thompson et al., 2024) to estimate surface reflectance spectra (Coleman et al., 2024; Green, 2022). Minerals are identified from reflectance features using the linear feature matching algorithm Tetracorder at the instrument's native 60-m spatial sampling (Clark et al., 2003, 2024). EMIT scenes are mosaiced together by selecting cloud-free measurements with the minimum solar zenith angle from between August 2022 and October 2024. Locations with less than 65% bare soil are excluded, along with areas with consolidated mineralogy (e.g., rock formations), and all pixels within each half-degree grid cell are averaged together to identify the mass fraction. The EMIT mineral maps do not acquire data poleward of  $\pm 52^\circ$  latitude and these regions instead use data from previous mineral mass fraction estimates (Claquin et al., 1999; Journet et al., 2014). While EMIT maps represent remote sensing measurements, they do include errors. For example, Ca (predominately from calcite) may be underreported by the EMIT data due to detection issues (Clark et al., 2024). The diagnostic absorption for carbonates is at 2.3 microns and the absorption coefficient in that spectral region interferes with the corresponding absorption coefficients for phyllosilicates, including muscovite and illite. Thus the presence of significant muscovite and illite can mask the carbonate absorptions, leading to carbonate non-detections or lower detections. A second effect has to do with the derived reflectance calibration in the EMIT data. Some, but not all, scenes have an artifact near 2.3 microns that can affect detection of minerals with 2.3

micron absorptions, including calcite, dolomite, chlorites, and serpentines, again leading to either non or lower detections (Clark et al., 2024). Ongoing work is attempting to remove these issues from the EMIT product.

EMIT does not directly observe quartz and feldspar. These abundance maps are produced using the calibrated VSWIR reflectance spectra interpreted through a Hapke-style model of the soil surface coupled with the Tetraformer spectral-matching algorithm (Clark et al., 2024). Size-resolved mineral mass fractions in aerosols at emission are estimated based on Brittle Fragmentation Theory (Kok, 2011) after partitioning the EMIT mineral bulk abundances into clay ( $\leq 2 \mu\text{m}$ ) and silt ( $2\text{--}50 \mu\text{m}$ ) fractions based on empirical relationships that incorporate soil texture data, Food and Agriculture Organization (FAO) soil classifications, and additional observational constraints (Claquin et al., 1999; Journet et al., 2014). For this study iron oxides are assumed to be evenly distributed across particle sizes, although there is evidence that there may be more iron oxides in smaller sizes (Panta et al., 2023). Iron oxides consist of hematite and goethite following Li et al. (2022). There is limited availability of optical properties for chlorite and vermiculite, and there is very little in the inventories, and thus these two minerals are combined into the kaolinite for this study.

### 2.3. Other Aerosol Sources of Al, Ca, Fe, Si, and Ti

The model was modified to allow the addition of several new aerosol sources using the speciated dust version (Li et al., 2021, 2024), that are simulated separately and added together to get the resulting distribution and deposition. The additional sources of aerosols (such as industrial ash and primary biogenic particles) use the same chemical and optical properties as bulk dust (e.g., density, shape and complex refractive index).

Industrial emissions include metallic aerosol particles (e.g., industrial ash) from combustion, and we base our emissions here on the detailed study of mineralogy from combustion focused on Fe (Rathod et al., 2020). That study provides spatial distributions of the fine and coarse estimates of Fe from coal, wood, oil, gasoline, diesel, smelting, wood fuels and fires for Fe (Rathod et al., 2020). The ratios of Al, Ca, Si, and Ti to Fe are estimated from literature sources and used to simulate the combustion contribution to these elements (Table S2 in Supporting Information S1). Road dust emissions estimated from the GAIN model are added to the model using the ECLIPSEV6\_CLE base case (Klimont et al., 2017; Philip et al., 2017). Emissions from cement production due to combustion (of whatever fuel is used) are included here, but not from fugitive emissions from cement plants (Rathod et al., 2020). Biomass burning emissions are included using emissions of black carbon multiplied by literature derived ratios (Table S3 in Supporting Information S1). We are missing possible braking emissions.

Previous estimates show natural primary biogenic particles as sources of these elements (Mahowald et al., 2018; Rauch & Pacyna, 2009). Primary biogenic particles are accidentally or deliberately released from ecosystems either as whole particles, such as bacteria, pollen, or spores, or as entrained leaf or insect pieces (Burrows et al., 2009; Despres et al., 2012; Heald & Spracklen, 2009; Jaenicke, 2005; Mahowald et al., 2008) and here we use an identical approach as in Mahowald, Li, Vira, et al. (2025). Assumptions about size are likely to be very important for the resulting distribution and impacts. For example, studies show that P budgets are quite different if 5 different size bins or 1 size bin are included in models (Brahney et al., 2015). Bacteria sources are included from a monthly climatology (Burrows et al., 2009). Fungal spore emissions are calculated offline on observed leaf area index, temperature, and a source parameterization (Heald & Spracklen, 2009; Janssen et al., 2020). An additional terrestrial biogenic source is estimated based on leaf area index following Mahowald et al. (2008). Elemental composition of primary biogenic materials is estimated from the limited literature (Table S3 in Supporting Information S1).

Sea spray contains trace quantities of metals (Marsay et al., 2022; Rauch & Pacyna, 2009). Based on the available literature, the fraction of each element from these sources is multiplied by the sea salt source in the model (Table S3 in Supporting Information S1). Note that it is possible that these studies include the impact of marine primary biogenic material and thus no additional marine primary biogenic particles are included.

Studies have shown that volcanoes can be an important contributor to trace elements in aerosols, through eruptive activities and degassing (Mahowald et al., 2018; Rauch & Pacyna, 2009). While most metals are likely to come from eruptive activities, there is no climatological database of ash currently available (Hornby et al., 2023; Rauch & Pacyna, 2009), thus we use a non-eruptive source spatial distribution for sulfur (S) for this study (Spiro et al., 1992), with a constant source across the time periods. For volcanic sources, the concentration of trace elements is commonly expressed using their ratio to S. The amount of Fe emission for this study is tuned to be

close to that in Rauch & Pacyna, 2009 at a global level, and the ratios of the elements are used to deduce the emissions of other elements. We summarize recently measured concentrations of the elements from volcanic ash (Table S4 in Supporting Information S1). The amount of different elements is sensitive to the size fraction (Tomašek et al., 2025) as well as the type of volcano (Hornby et al., 2023): the variability due to the type of volcano and composition appears stronger than the size differences (López-Darias et al., 2025; Tomašek et al., 2025). The size is especially important for the Si content, as there is a strong size segregation for Si, so we reduce the Si in the PM<sub>10</sub> size fraction assumed here, but otherwise use the Hornby et al., 2023 estimates. We note that this approach does not rely on a direct physical conversion factor between sulfur emissions and ash, but instead uses observed elemental-to-sulfur ratios and global constraints to approximate emissions. This introduces substantial uncertainty, which if volcanoes are important, should be refined in the future.

The total emissions from all sources is listed in Table S5 in Supporting Information S1 and discussed in more detail in Section 3.2. The surface concentration fraction that is in the PM<sub>2.5</sub> size fraction is shown for each source and each element in Table S5 in Supporting Information S1, the remaining fraction largely represents coarse-mode (PM<sub>2.5</sub>-PM<sub>10</sub>) aerosol, as the model primarily resolves particles within the PM<sub>10</sub> range.

#### 2.4. Atmospheric Surface Concentration Observations

Elemental in situ concentration measurements are routinely made in several global networks, and these are combined with field studies and cruises to create a global distribution of the different aerosols in atmospheric surface concentrations as part of the COARSEMAP/AEROMAP effort (Lu et al., 2024; Wiedinmyer et al., 2017; Wong et al., 2021). Data from land-based stations are collected by announcing the effort publicly (Wiedinmyer et al., 2017), searching the literature for sources, individually contacting authors, as well as downloading publicly-available data (e.g., <https://cbas-data.nilu.no/>; <https://www.spartan-network.org/>; <https://www.epa.gov/outdoor-air-quality-data>). Since one of our goals is to constrain atmospheric deposition to oceans, we also include cruise data (e.g., <https://www.geotraces.org/>; Baker et al., 2013). We focus here on total elemental amounts, not on the soluble fraction, for which there is much less data.

For land-based data, we focus on the climatological annual means for 1986–2023 which are calculated for all values at each station that are above the detection limits. At some stations or times, concentrations can be below the detection limit, and excluding these data or time periods could bias our average values. We focus only on the stations that have more than 50% of the data above the detection limit. For those included stations, if the values were reported as below the detection limit, we include in the average one-third of the minimum detection limit. The reported detection limits should provide an upper limit of aerosol mass and allow us to include sites whose observations are otherwise too low to include, while reducing the potential biasing of our compilation toward higher values (Data Set S1). Studies have suggested errors of up to 30% in coarse mode detection of Al and Si, with smaller errors in PM<sub>2.5</sub> (Hyslop et al., 2015; Liu et al., 2025; Lucarelli et al., 2018).

The largest variability in surface concentration aerosol distributions at the global scale is the spatial distribution of the annual mean (Mahowald, Li, Vira, et al., 2025), so we focus on understanding and reproducing the climatological mean surface concentration spatial distribution in the PM<sub>10</sub>, and consider the PM<sub>2.5</sub> aerosol in the supplement. Variability within 1 year at the same land-based station, between different years at the same station, spatial variability within one model grid box due to the coarse spatial resolution, measurement uncertainties and differences in measurement methods, all contribute to uncertainty in these comparisons (Mahowald, Li, Vira, et al., 2025). Although episodic events may lead to larger deviations, we estimate that at land-based stations model values within a factor of 3 of the observed values should be considered within the uncertainties (Mahowald, Li, Vira, et al., 2025). This is much less than spatial distributions which vary over 4–5 orders of magnitude. Comparisons are made to the surface layer in the model (bottom ~50 m), and thus the results are sensitive to errors in the boundary layer processes in the models.

In dust dominated regions (as defined by the model as dust is more than 50% of the total) we look at the relative amount of different elements compared to Al to evaluate the composition of the dust. For these comparisons, the errors from spatial and temporal variability become less important, and we assume the largest errors are due to measurement errors. Because Al content in dust is relatively constant, it is usually used as the standard, although Al and Si may have 30% errors due to measurement methods as discussed above. Propagating these errors in the denominator and numerator produces error bars of a factor of 2, which are used for the comparisons for different

elements over Al. Notice that our definition of dust dominated may be biased due to errors in the sources or transport of aerosols.

For the ocean-based data, we include all measurements across all time periods, although these are limited. Since many measurements represent just one daily or weekly average, these are more likely to be subject to the large temporal variability and less representative of the annual mean. Previous studies have estimated that the uncertainty in model (annual average) versus single observation is close to one order of magnitude, and we use that here for comparisons (Mahowald et al., 2008).

Annual average and climatological monthly mean values for the station data are provided in the associated Zenodo database (Mahowald, Li, Vira, et al., 2025), as well as in Data Set S1. Compiled cruise data are provided in Data Set S2. Information about location and the relevant citations are included in these data sets.

## 2.5. Model/Data Comparisons

We focus this study on the PM<sub>10</sub> component, as that is the relevant size fraction for biogeochemistry (since most of the mass is in the larger size fractions; Mahowald et al., 2011), and PM<sub>2.5</sub> data are shown in the supplement. Comparisons are made at the global and regional level between the annual averages in the model versus the observations, to see how the model performs in simulating large spatial gradients, using the regions defined in Figure S2 in Supporting Information S1 (similar to Mahowald, Li, Vira, et al. (2025)). Because in some regions there are too many data, obscuring individual points, we average the observations to a 2° × 2° grid to better discern differences between model values and observations. The grid used is sampled using every other model grid point.

Statistical comparisons include the correlation coefficients (Kendall ranked correlation), slope of the observations versus the model with uncertainty, root mean squared differences between model and observations, average observational values, average model values (at the observational sites), the fraction of comparisons when the model and observations are within the uncertainty bounds discussed in Section 2.4, and the number of observations. These statistics are included in supplemental tables discussed in the text.

## 2.6. Preindustrial Versus Present Day Estimates

The descriptions above are for the present day (model is for 2013–2015; observations are 1980s–2025). To estimate preindustrial values, we assume that all the anthropogenic sources are zero, and non-dust natural sources are the same as present day (e.g., volcanoes, sea salts and primary biogenic particles). As these sources tend to be smaller, these assumptions are unlikely to significantly impact our results. For preindustrial dust sources, we use the paleodata from Kok et al. (2023) which indicates that dust has changed about 55% since preindustrial times. We force a change in the emissions at each grid box in each region so that the mean will be consistent with the paleodata.

## 3. Results and Discussion

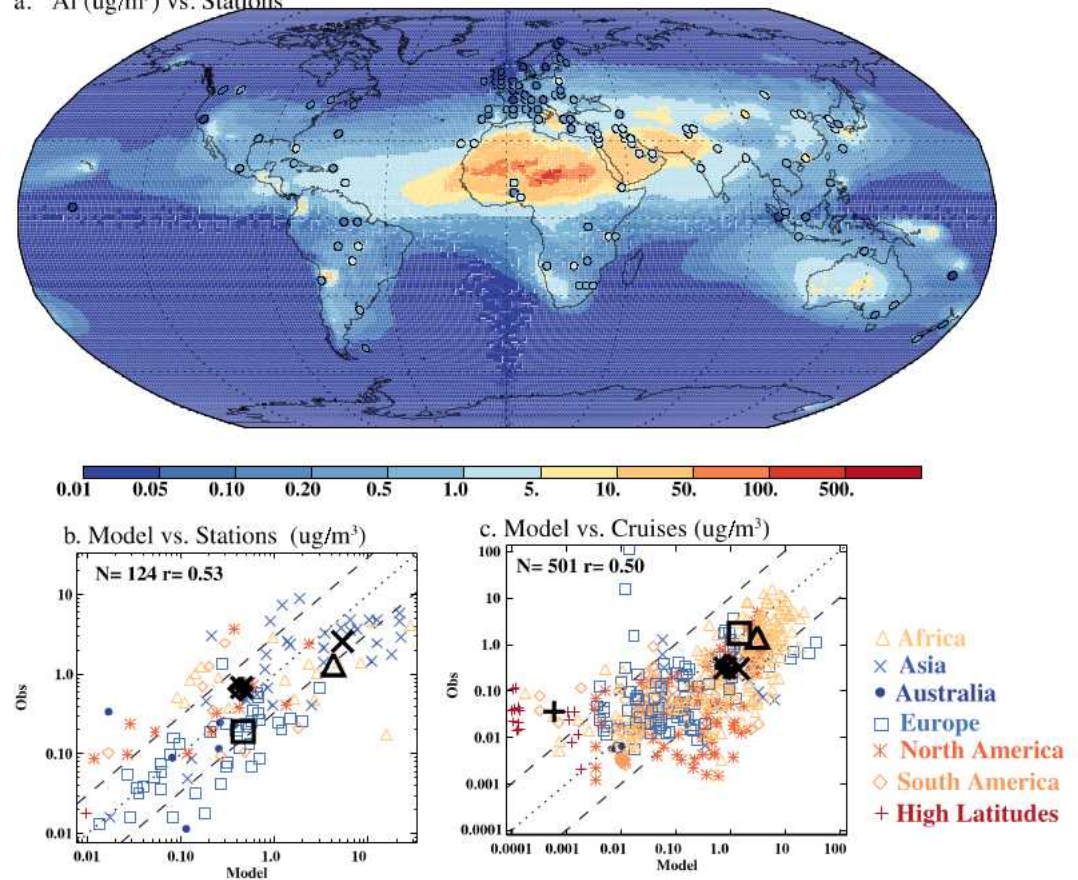
Since this research represents the first study of multiple sources, not just of dust, but also of Al, Ca, Si, and Ti, we start with an evaluation of the atmospheric concentrations resulting from the assumptions described in Section 2. For Fe, this is less important, as it was previously evaluated (e.g., Hamilton et al., 2019; Mahowald et al., 2009), and we use those examples to contextualize the new results from this study.

### 3.1. Model/Data Comparisons

#### 3.1.1. Aluminum (Al)

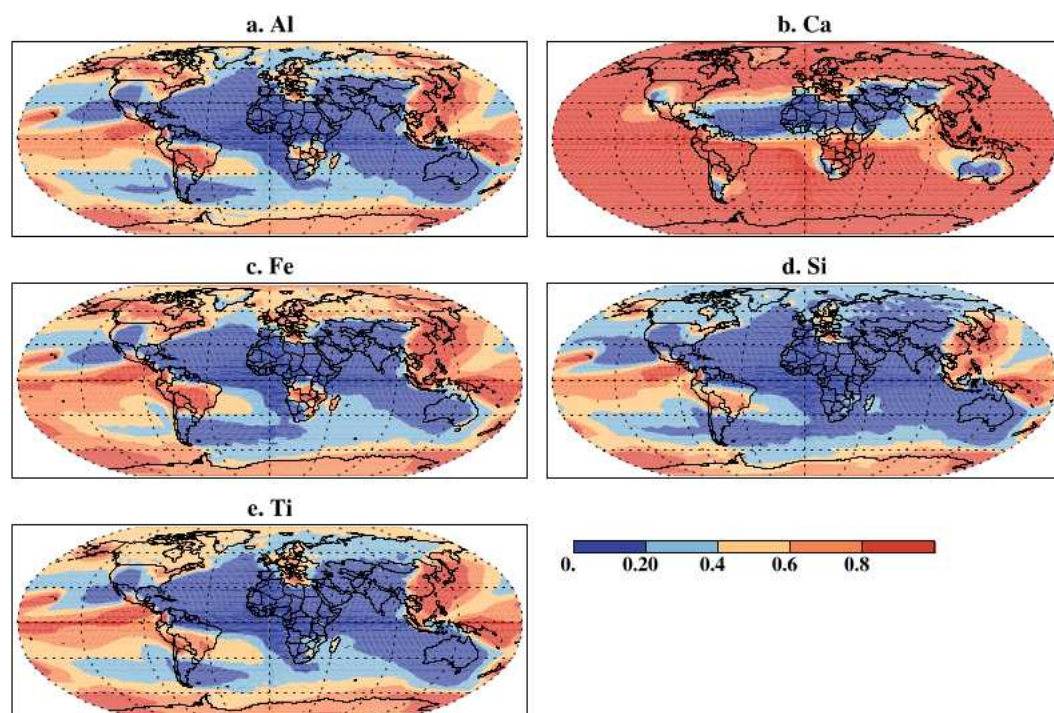
The spatial distribution of the modeled PM<sub>10</sub> surface concentrations of Al has the maximum in the dust belt, across North Africa and into the Middle East and Central Asia (Figure 1; regions are defined in Figure S2 in Supporting Information S1). This feature is as expected and will be true of all the elements in this paper (fraction shown explicitly in Figure 2) since Al, Ca, Fe, Si, and Ti all are major constituents of desert dust, which is the most abundant aerosol by mass. Land-based station data are limited for Al in PM<sub>10</sub> (Figure 1a), particularly in the desert regions, and they broadly agree with the model across four orders of magnitude. Some of the highest concentrations are in eastern Asia near Shanghai (Figure 1), where there may be significant non-desert dust sources, such

a. Al ( $\mu\text{g}/\text{m}^3$ ) vs. Stations



**Figure 1.** Al surface concentrations in the  $\text{PM}_{10}$  size fraction. Modeled values in  $\mu\text{g}/\text{m}^3$  are shown in the background in (a) while the circles represent the observational measurements from land-based stations using the same color bar. A comparison of the model (x-axis) to the observations (y-axis) is shown for the station-based land data (b) and the cruise data (c). The spatial comparison between the model and cruise data is shown in Figure S4a in Supporting Information S1. For clarity, all the observations are gridded to  $2^\circ \times 2^\circ$  cells. In the scatter plots, the color and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are the factor of 3 for land-based measurements (b) and 10 for ocean cruise data (c) uncertainty estimates. More statistics are shown in Table S6 in Supporting Information S1. The regions are defined in Figure S2 in Supporting Information S1. More detailed views of the regions are shown in Figure S6 in Supporting Information S1.  $N$  and  $r$  represent the number of observations and correlation coefficient, respectively.

as agricultural, construction, or road dust. The model tends to overpredict Al over Africa and Asia which may be due to an overprediction of dust or a mistake in the seasonality (Formenti et al., 2014) in the model in those regions (e.g., Aboagye-Okyere et al., 2025), although the regional averages are just within the uncertainty bounds. However, we do not see this overprediction consistently across all the dust-derived elements (below) nor in the dust specific comparisons (Figure S1 in Supporting Information S1), so it could also be an error in the amount of Al in the dust, or due to other emissions, since our observations of Al are in regions where non-dust sources also contribute (Figure 2). In addition, it could be due to an underestimate in assessment of the Si and Al in the coarse particles when measured with techniques based on the detection of emitted X-rays such as Particle Induced X-ray Emission (PIXE) and X-Ray Fluorescence (XRF) (Lucarelli et al., 2018); this effect is due to the self-absorption of the low-energy X-rays inside the particle and may cause an underestimation of the order of 20% if not properly corrected (Formenti et al., 2010). Note that it is difficult to match the aerosol optical depth, deposition records and surface concentrations close to dust source regions at the same time as is the case in most dust models (Albani et al., 2014; Huneus et al., 2011), perhaps because of errors due to not accurately including larger dust particles (Li et al., 2026). Because we are most interested in biogeochemical effects, the match in dust deposition (Figure S1c in Supporting Information S1) is most important.



**Figure 2.** Non-dust fraction for each of the elements. Model based estimates of the annual average concentration contribution from non-dust sources for (a) Al, (b) Ca, (c) Fe, (d) Si, and (e) Ti.

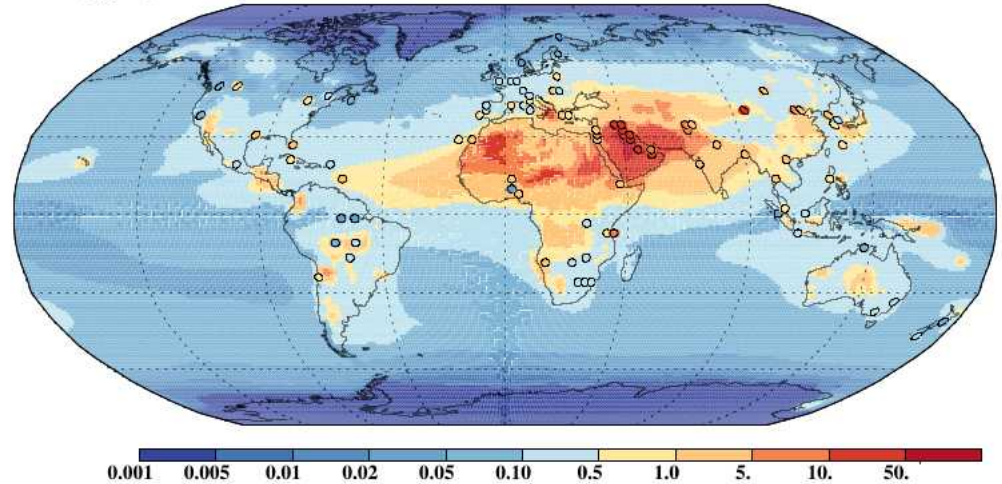
The dominant dust sources of Al are the minerals Illite and Kaolinite (Figure S7 in Supporting Information S1). Non-dust Al sources dominate away from the main dust sources areas (Figure 2), and the dominant sources spatially are volcanoes over most of the oceans, fires and coal burning over some land regions (Figure S8 in Supporting Information S1; budgets will be discussed in Section 3.2). The simulation of Al in the non-dust dominated regions like North America and Europe, with abundant anthropogenic aerosol sources (Figure 2), suggests that the non-dust sources of Al are roughly correct (North America) or slightly too high (Europe) (Figures 1a and 1b). The  $PM_{2.5}$  simulations of Al are better across most regions, except for Europe (Figures S2a and S2b in Supporting Information S1), and these smaller sizes will tend to be more dominated by anthropogenic emissions. The percent of the observations and model comparisons that are outside of the uncertainty bounds of  $3\times$  (Section 2.4) is 47% and 28% for  $PM_{10}$  and  $PM_{2.5}$  (Tables S6 and S7 in Supporting Information S1), respectively; for  $PM_{10}$  these are larger than expected if we consider these 1-sigma bounds, suggesting improved emission estimates for  $PM_{10}$  are needed.

The cruise data for Al in the  $PM_{10}$  size fraction show more correspondence with the average from the observations than the land station data, except in high latitudes (Figure S4a in Supporting Information S1 and Figure 1c). Notice that there are very high values from cruise observations close to Iceland, consistent with an episodic volcanic source that we are not simulating (Alex Baker, personal communication, 7 March 2024; Achterberg et al., 2021). There are much less cruise data for the  $PM_{2.5}$  size fraction, and they show that the regional averages are within the uncertainty bounds, although the model overpredicted observations at many sites. (Figures S5a and S5b; Table S9 in Supporting Information S1).

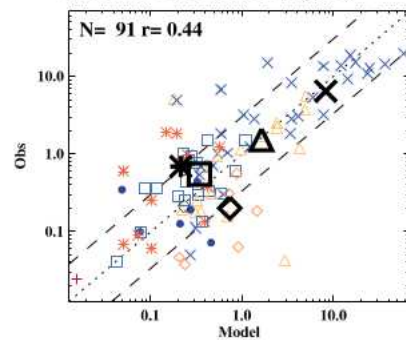
### 3.1.2. Calcium (Ca)

For Ca, the maximum modeled concentrations are again in desert regions, and are verified to a limited extent by  $PM_{10}$  land-based observations (Figures 3a and 3b). The model does better simulating Ca than Al across Asia, Africa, and Europe, but not South America where the modeled values are too high, and North America where the modeled values tend to be too low (Figure 3b vs. Figure 1b). In non-dust dominated regions, such as parts of Europe or North America which have good data coverage (Figures 2d and 3a), the model is similar to available observations, suggesting that the non-dust sources in the model are reasonably represented within observational

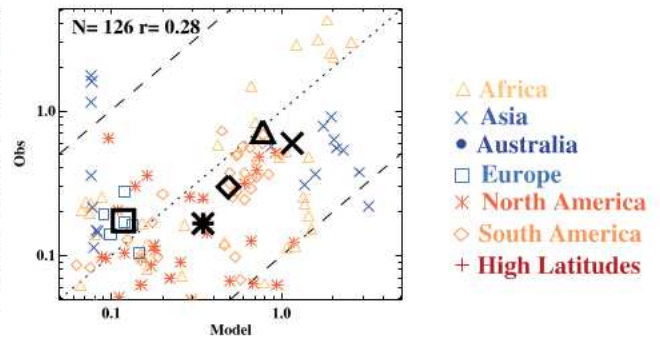
a. Ca ( $\mu\text{g}/\text{m}^3$ ) vs. Stations



b. Model vs. Stations ( $\mu\text{g}/\text{m}^3$ )



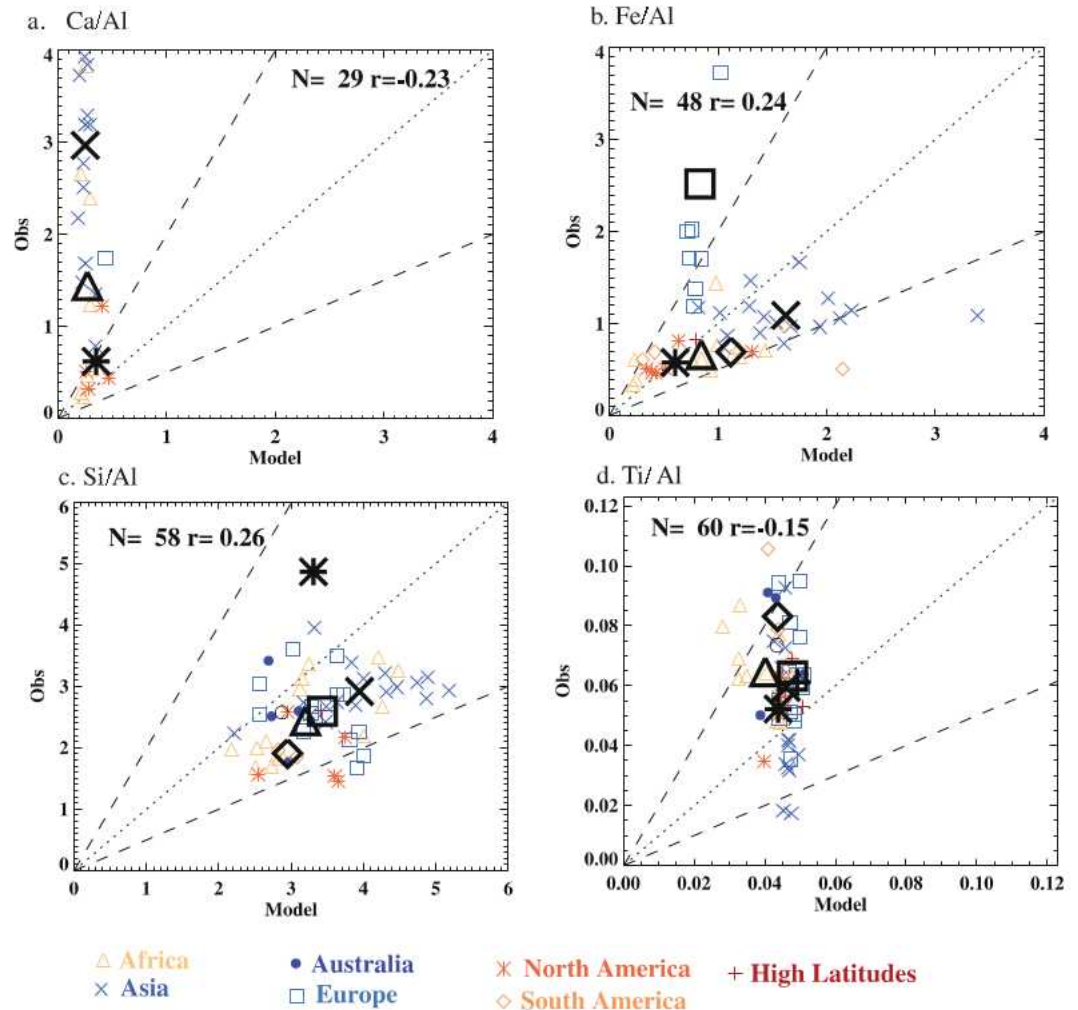
c. Model vs. Cruises ( $\mu\text{g}/\text{m}^3$ )



**Figure 3.** Same as Figure 1 but for Ca surface concentrations in the  $\text{PM}_{10}$  size fraction. Modeled values in  $\mu\text{g}/\text{m}^3$  are shown in the background in (a), while the circles represent the observational measurements from land-based stations using the same color bar. A comparison of the model ( $x$ -axis) to the observations ( $y$ -axis) is shown for the station-based data (b) and the cruise data (c). The spatial comparison between the model and cruise data is shown in Figure S4b in Supporting Information S1. For clarity, all the observations are gridded to  $2^\circ \times 2^\circ$  cells. In the scatter plots, the color and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are the factor of 3 for land-based measurements (b) and 10 for ocean cruise data (c) uncertainty estimates. More statistics are shown in Table S6 in Supporting Information S1. More detailed views of the regions are shown in Figure S9 in Supporting Information S1.  $N$  and  $r$  represent the number of observations and correlation coefficient, respectively.

uncertainty. The dominant dust source comes from Calcite (Figure S11 in Supporting Information S1) while non-dust sources of Ca are modeled to be sea salts over the oceans, and volcanoes, fires and coal over some land regions (Figure S12 in Supporting Information S1). Sources such as dust from agriculture or construction are not included in the model. Globally 43% and 47% of the observational/model comparison were outside the  $3\times$  uncertainty bounds (Tables S6 and S7 in Supporting Information S1) for  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$ , respectively: so the model does better for predicting Ca in  $\text{PM}_{10}$  than for  $\text{PM}_{2.5}$  in contrast to the results for Al. Model results for  $\text{PM}_{2.5}$  across South America are especially high, but results in the other regions are within the uncertainties, although the model produces values that are a little high for Africa and North America (Figures S3c and S3d in Supporting Information S1).

If we focus on the ratios of Ca/Al in dust dominated regions (Figure 4a), we see that the model is underpredicting the ratio of Ca/Al. This is likely due to the under-detection of calcite in the EMIT methods (see methods; Clark et al., 2024), although some of it could be due to under-detection of Al in the  $\text{PM}_{10}$  atmospheric concentrations (see methods; Lucarelli et al., 2018; Hyslop et al., 2015; Liu et al., 2025). The underprediction by the model is the



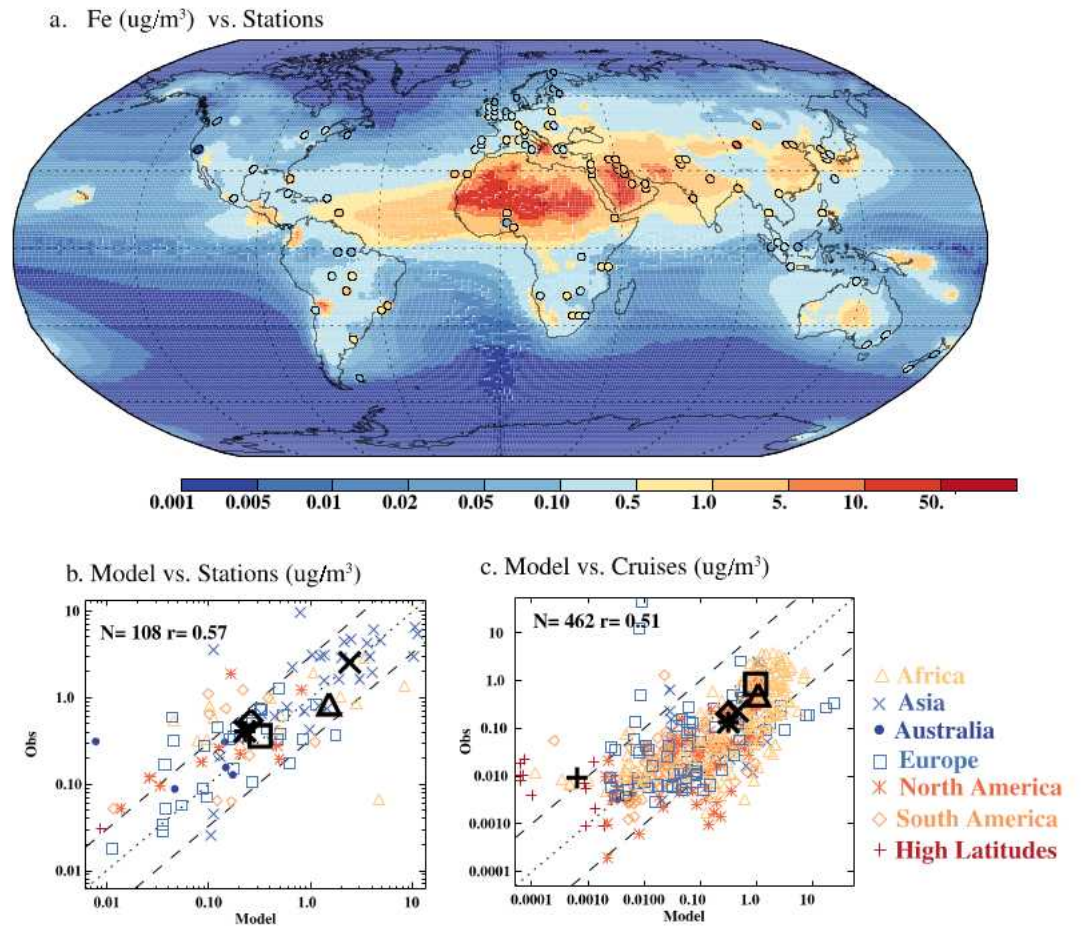
**Figure 4.** Scatter plot of the  $PM_{10}$  elemental ratios between model (x-axis) and observations (y-axis) for (a) Ca/Al, (b) Fe/Al, (c) Si/Al, and (d) Ti/Al where the ratios of concentrations are in ( $\mu\text{g}/\text{m}^3$  divided by  $\mu\text{g}/\text{m}^3$ ). In the scatter plots, the color and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are the factor of 2 uncertainties bound as described in the methods section. The spatial maps of the ratios are available in Figure S10 in Supporting Information S1.

worst in Asia and is seen to some extent in Africa. Improved methods to detect Ca derived minerals in EMIT are important to improve Ca simulations.

The cruise data suggest that the Ca in  $PM_{10}$  is similar for the model and the observations (Figure 3c; Table S8 in Supporting Information S1). Similarly, the limited  $PM_{2.5}$  cruise data also suggests that the model is within the uncertainties of the model-data comparison for most of the observations (Figures S5c and S5d; Table S9 in Supporting Information S1).

### 3.1.3. Iron (Fe)

Simulated Fe was more highly correlated with available data compared to the other elements (Figure 5a vs. Figures 1a and 3a). This is consistent with its emissions inventory and transport modeling being much more mature than for other elements, and the emission data set we use here being focused on simulating Fe (Hand et al., 2004; Myriokefalitakis et al., 2018; Rathod et al., 2020, 2024; Scanza et al., 2018). The averages across the regions are within the 3 $\times$  uncertainty, and the concentrations in non-natural dust dominated regions are similar between the model and observations (Figure 5). Studies have shown that the model is less able to simulate natural Fe derived from dust than anthropogenic sources of Fe (Rathod et al., 2020), but that anthropogenic sources are

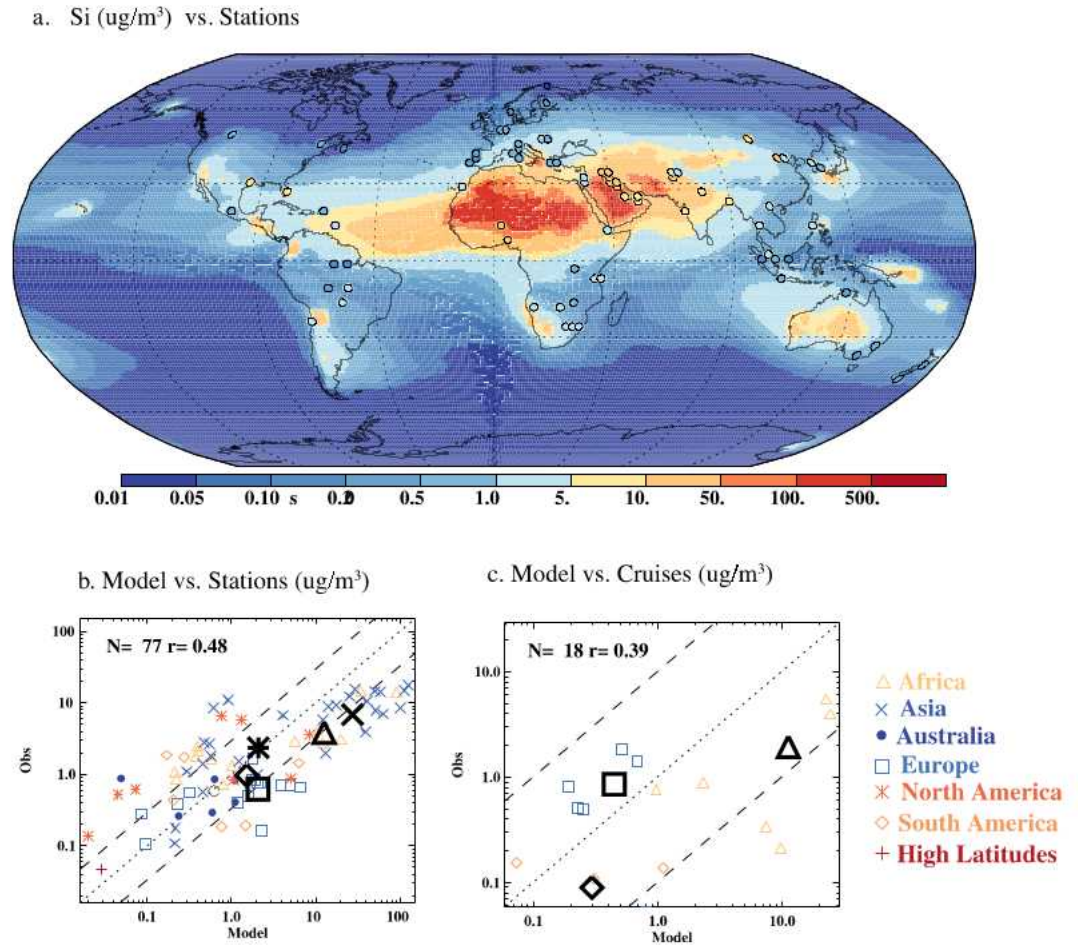


**Figure 5.** Same as Figure 1 but for Fe surface concentrations in the  $\text{PM}_{10}$  size fraction. Modeled values in  $\mu\text{g}/\text{m}^3$  are shown in the background in (a), while the circles represent the observational measurements from land-based stations using the same color bar. A comparison of the model ( $x$ -axis) to the observations ( $y$ -axis) is shown for the station-based data (b) and the cruise data (c). The spatial comparison between the model and cruise data is shown in Figure S4c in Supporting Information S1. For clarity, all the observations are gridded to a  $2^\circ \times 2^\circ$  degrees. In the scatter plots, the color and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are the factor of 3 for land-based measurements (b) and 10 for ocean cruise data (c) uncertainty estimates. More statistics are shown in Table S6 in Supporting Information S1. More detailed views of the regions are shown in Figure S13 in Supporting Information S1.  $N$  and  $r$  represent the number of observations and correlation coefficient, respectively.

within a factor of 3 (Rathod et al., 2024). This suggests that difficulties in capturing the natural sources of dust derive from issues in simulating the elemental composition. The model suggests that the dominant dust sources of Fe are iron oxides (Figure S14 in Supporting Information S1) and of non-dust sources are volcanoes over the oceans, and fires and non-Al smelting over land regions (Figure S15 in Supporting Information S1). Comparisons of the Fe/Al ratios in dust dominated regions (Figure 4b) show that in most dust regions, the Fe/Al ratio is well simulated, except in Europe. The threshold used here (50% dust in the model) may not be strict enough to be able to filter out anthropogenic Fe sources (e.g., combustion).

Simulations of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  Fe are mostly within the uncertainties, especially for regional averages, although the results are a little high for South America (Figures S3e and S3f in Supporting Information S1). 37% and 24% of the  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  model/data comparisons are outside  $3\times$  uncertainty (Tables S6 and S7 in Supporting Information S1).

Cruise data show that the average model results in different regions are close to averaged observations for  $\text{PM}_{10}$  (Figures S4c and 5c; Table S8 in Supporting Information S1) and  $\text{PM}_{2.5}$  (Figures S5e and S5f; Table S9 in



**Figure 6.** Same as Figure 1 but for Si surface concentrations in the  $\text{PM}_{10}$  size fraction. Modeled values in  $\mu\text{g}/\text{m}^3$  are shown in the background in (a), while the circles represent the observational measurements from land-based stations using the same color bar. A comparison of the model (x-axis) to the observations (y-axis) is shown for the station-based data (b) and the cruise data (c). The spatial comparison between the model and cruise data is shown in Figure S4d in Supporting Information S1. For clarity, all the observations are gridded to a  $2^\circ \times 2^\circ$  degrees. In the scatter plots, the color and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are the factor of 3 for land-based measurements (b) and 10 for ocean cruise data (c) uncertainty estimates. More statistics are shown in Table S6 in Supporting Information S1. More detailed views of the regions are shown in Figure S17 in Supporting Information S1.  $N$  and  $r$  represent the number of observations and correlation coefficient, respectively.

Supporting Information S1), similar to previous results with a previous version of this model (Hamilton et al., 2019). For iron, there is another compilation of cruise data from Myriokefalitakis et al., 2018, and this model simulation gives a similar comparison to the cruise data as the previous study (Figure 5c; Figures S4c and S16 in Supporting Information S1). Further studies will consider in more detail the impact on soluble iron of the EMIT data set.

### 3.1.4. Silicon (Si)

Model simulations of Si show a maximum over desert regions, but also near volcanic regions (e.g., Sicily, Indonesia, and Japan) (Figure 6a). Model simulations of  $\text{PM}_{10}$  Si tend to be higher than the available station observations in the regional means, especially in Europe, Africa, and Asia (Figures 6a and 6b). This is consistent with Al, in suggesting dust sources are very high in Africa, but inconsistent with the results from Ca and Fe. The dominant dust source of Si is Quartz (Figure S18 in Supporting Information S1). The dominant non-dust source of Si in the model is volcanoes over the ocean with some contribution from coal over some land regions (Figure S19

in Supporting Information S1). In non-dust dominated regions (Figure 2d), such as parts of Europe or North America which have good data coverage, the model is similar to available observations, suggesting that the non-dust sources in the model are approximately correct. Volcanic sources of silicon may be poorly represented, as the model shows a maximum over Sicily from volcanoes (Figure 6a). The representation of volcanoes in these simulations is very simple and needs improvement, since it assumes constant outgassing, instead of intermittent ash generation. In dust-dominated regions, comparisons show a good correspondence of the ratios of Si/Al between model and data (Figure 4c).

Simulations of  $PM_{2.5}$  Si over land tend to be within the 3 $\times$  uncertainty bars, but slightly high (Figures S3g and S3h in Supporting Information S1). Approximately 51% and 22% of the Si observation-model comparisons for  $PM_{10}$  and  $PM_{2.5}$ , respectively, are outside the uncertainty bounds (Tables S6 and S7 in Supporting Information S1), suggesting that  $PM_{2.5}$  is better simulated than  $PM_{10}$ .

Si data are sparse in cruises, and what little data there are displays some correspondence with the simulations for  $PM_{10}$  (Figure S4d in Supporting Information S1 and Figure 5c; Table S8 in Supporting Information S1; there are no Si cruise data in the  $PM_{2.5}$  size fraction).

### 3.1.5. Titanium (Ti)

In the model simulations, the largest concentrations of Ti are near desert regions, or volcanoes, similar to Si and these are difficult to verify with the available observations (Figure 7). Over 4 orders of magnitude the model simulates much of the spatial variability (Figures 7a and 7c) with the regional averages all within the uncertainty bounds for  $PM_{10}$ . Ti is a trace amount in dust, and comes predominately associated with Illite and Kaolinite (Figure S21 in Supporting Information S1). The non-dust dominated sources of Ti over oceans (and some land regions) is volcanoes in the model, and over land, coal (Figure S22 in Supporting Information S1). In dust dominated regions, the Ti/Al ratio in the model is much less variable than in the observations (Figure 4d), but the model is just within the error bars in all regions.

For  $PM_{2.5}$ , the model tends to overpredict the regional averages in Europe and Asia (Figures S3i and S3j in Supporting Information S1). Approximately 41% and 27% of the observation-model comparisons for  $PM_{10}$  and  $PM_{2.5}$ , respectively, are within the uncertainty bounds (Tables S6 and S7 in Supporting Information S1), which suggests the  $PM_{10}$  simulations have more difficulty simulating the spatial spread in the values than the  $PM_{2.5}$ .

The cruise data suggests that the regional averages are similar between the model and observations, as the regional means are within the uncertainties in the  $PM_{10}$  size fraction (Figure 7c and Figure S4e; Table S8 in Supporting Information S1). This is not true in the high latitudes, as the Iceland volcano has been sampled in the observations but not in the model (Figure 7c and Figure S4e in Supporting Information S1). The limited  $PM_{2.5}$  cruise data suggests the model is within the uncertainties (Figures S5g and 5h; Table S9 in Supporting Information S1).

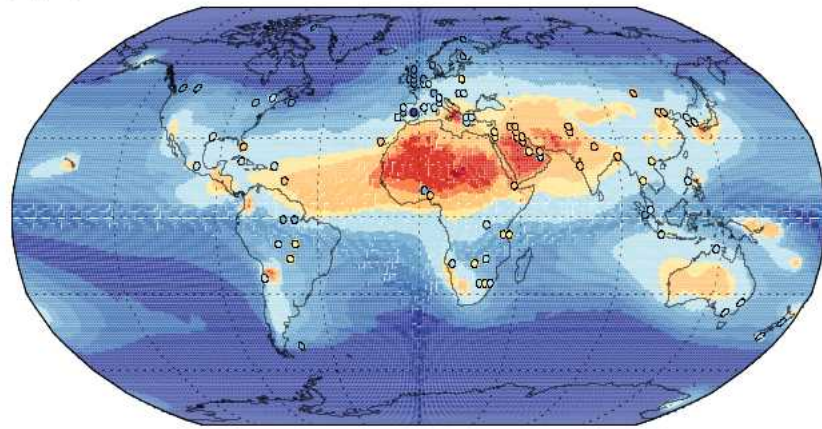
## 3.2. Elemental Budgets

The overall emissions of each element in the atmospheric  $PM_{10}$  size fraction vary from almost 1,000 Tg/year for Si to  $\sim$ 10 Tg/year for Ti (Figure 8a). There are only a few literature values for the budgets of these elements, but a recent review tried to summarize our understanding of Al, Fe, and Ti (Mahowald et al., 2018) based on existing literature (Nriagu, 1989; Nriagu & Pacyna, 1988; Pacyna & Pacyna, 2001; Schlesinger, 1997). Previous studies did not compare to available surface concentrations, and thus were not shown to match observational estimates. Al, Fe, and Ti budgets are slightly higher in this study compared to the earlier work (Mahowald et al., 2018). For these three metals, the largest contribution comes from dust, which is much better resolved in this study, since we use mineralogy based on space-borne data and consider transport.

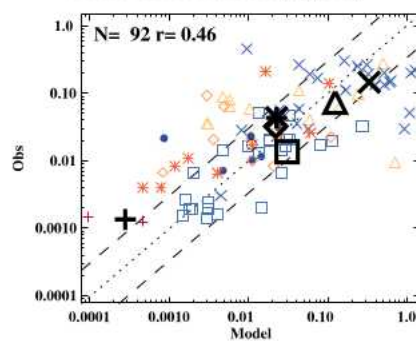
Within dust, the kaolinite fraction represents the most important sources for Al and Ti, while calcite is the most important for Ca (Figure 8b). Fe has roughly similar contributions from Fe oxides and illite, while Si comes mostly from quartz, but with significant contributions from illite (Figure 8b). Note that the observed Ca/Al ratios suggest the EMIT-based model is underestimating the Ca in dust (Figure 4a), and thus it is likely the global total from Calcite should be larger.

The contributions for all these elements are dominated by dust, as expected (Figure 8b: black and gray colors and patterns, Table 1). Contributions from anthropogenic sources tend to be small on a global average (pink and

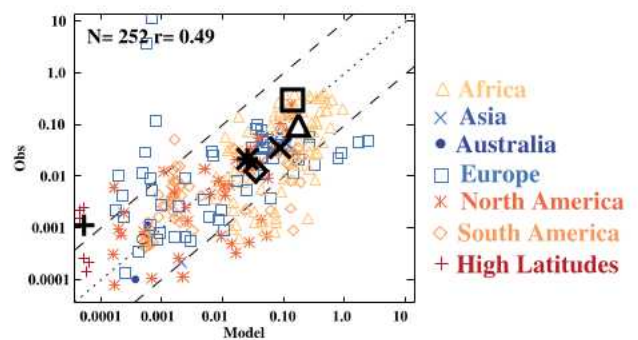
a. Ti ( $\mu\text{g}/\text{m}^3$ ) vs.Stations



b. Model vs. Stations ( $\mu\text{g}/\text{m}^3$ )



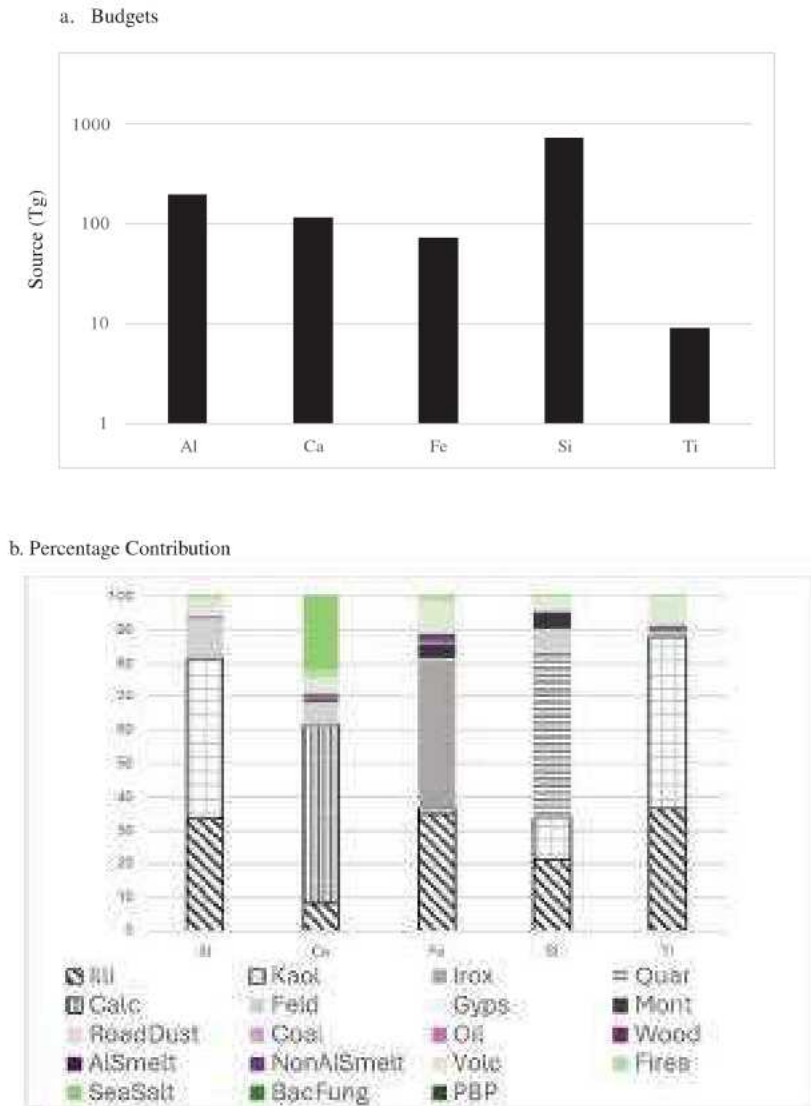
c. Model vs. Cruises ( $\mu\text{g}/\text{m}^3$ )



**Figure 7.** Same as Figure 1 but for Ti surface concentrations in the  $\text{PM}_{10}$  size fraction. Modeled values in  $\mu\text{g}/\text{m}^3$  are shown in the background in (a), while the circles represent the observational measurements from land-based stations using the same color bar. A comparison of the model (x-axis) to the observations (y-axis) is shown for the station-based data (b) and the cruise data (c). The spatial comparison between the model and cruise data is shown in Figure S4e in Supporting Information S1. For clarity, all the observations are gridded to a  $2^\circ \times 2^\circ$ . In the scatter plots, the color and symbols indicate the regions, the bold black symbols are the average across each region (indicated by the symbol), the dotted line is the 1:1 line and the dashed lines are the factor of 3 for land-based measurements (b) and 10 for ocean cruise data (c) uncertainty estimates. More statistics are shown in Table S6 in Supporting Information S1. More detailed views of the regions are shown in Figure S20 in Supporting Information S1.  $N$  and  $r$  represent the number of observations and correlation coefficient, respectively.

purple colors in Figure 8b, Table 1). Some sources were not included in this study, including dust from agricultural tillage. Other non-dust sources can be important for some elements, especially Ca (sea salts) and volcanoes are important for several of these elements (Figure 8b; Table 1). The volcanic source included in this estimate is not well parameterized, as it uses a distribution of emissions estimated from continuous outgassing of  $\text{SO}_2$ , which is then scaled to obtain estimates similar to previous studies (see Section 2.3). Since outgassing distributions and processes are quite different to the explosive eruptions that are likely to produce the insoluble ash-based aerosol elements included in our model, these estimates are likely incorrect. This analysis suggests further work is justified on ash-based aerosol elements in order to understand the role of explosive volcanoes in their budgets.

The overall contribution from general categories show that dust is the dominant source of these elements (Table 1), while volcanoes are the next largest source. Some very small contributions from anthropogenic, fires, sea salts, and biogenics are also seen (Table 1). Comparison with the limited previous studies suggests a similar importance of different sources (Table S10 in Supporting Information S1) for most elements. The tiny amounts of Al and Fe from sea spray and biogenic particles is smaller in this study than one previous estimate (Mahowald, Li,



**Figure 8.** Elemental budgets. The atmospheric global total emissions (Tg/year) in the PM<sub>10</sub> fraction is shown (a), with the contribution from each of the sources considered here in (b). The dust-based sources are in black and gray colors, while the pink and purple sources are anthropogenic and the green colors are non-dust natural aerosol sources. Emissions for each element for each source type is shown in Table S5 in Supporting Information S1. The abbreviations in the source contributions (b) are the following: Illi, Illite; Kaol, Kaolinite; Irox, Iron oxides; Quar, Quartz; Calc, Calcite; Feld, Feldspar; Gyps, Gypsum; Mont, Montmorillonite; RoadDust, Road dust; Coal, Coal burning; Oil, Oil burning; Wood, Wood burning; AlSmelt, Al smelting; NonAlSmelt, Non-Al smelting; Volc, Volcano; Fires, Open fires; SeaSalt, Sea Salts; BacFung, Bacteria and Fungi; PBP, other primary biogenic particles.

Vira, et al., 2025). Extraterrestrial sources of iron, from incoming meteorites for example, have been speculated to be important, but studies have shown their contribution is not significant even over remote ocean regions of soluble iron (Mahowald et al., 2009).

### 3.3. Deposition Changes Since Preindustrial Times

The extent of human modification of aerosol-induced deposition of important nutrients or pollutants is an important question for biogeochemists. Here we use the estimates from the model to look at the ratio of deposition for present day over preindustrial times, or how much deposition of each element is likely to have been changed. We assume that dust increased since preindustrial times, following the paleodata estimates in Kok et al. (2023), although these are uncertain, and based on limited data. These estimates suggest approximately a 55% increase in

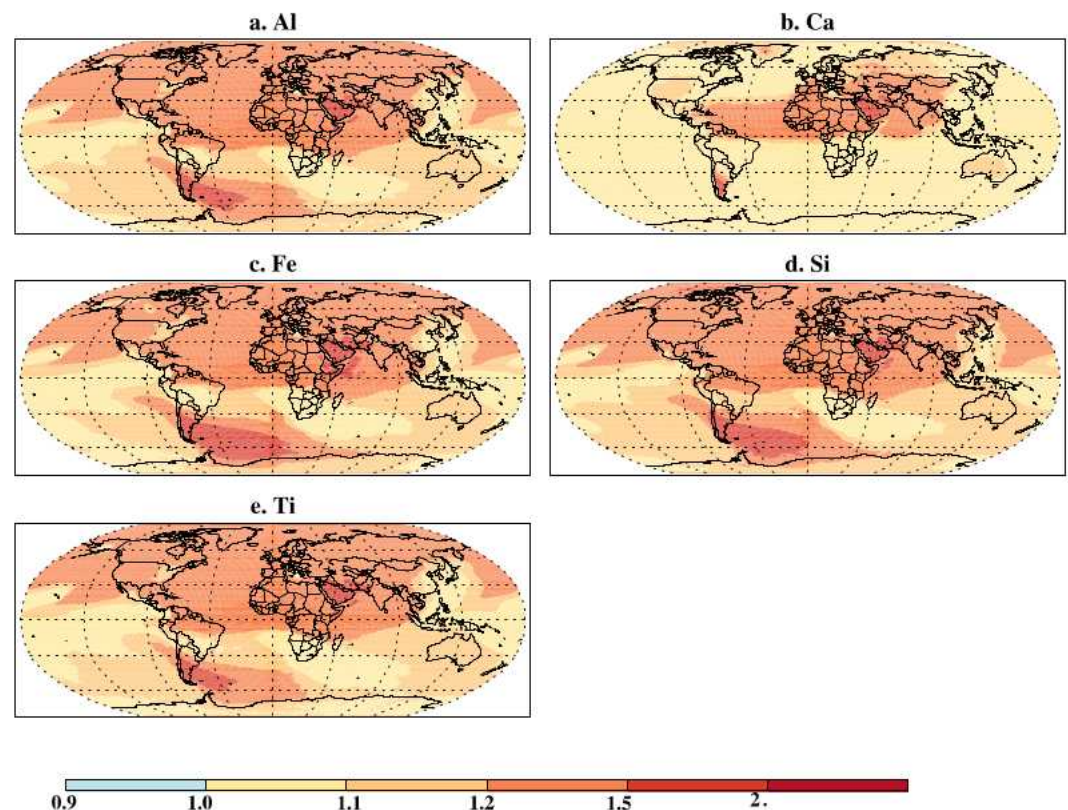
**Table 1**  
*Elemental Budgets*

	Al	Ca	Fe	Si	Ti
Total (Tg/year)	200	118	74	740	9
% Dust	93	69	86	95	91
% Anthropogenic	1	1	3	<1	1
% Fire	<1	2	1	<1	<1
% Biogenic	<1	<1	<1	<1	<1
% Sea Spray	<1	21	<1	<1	<1
% Volcanoes	6	5	10	5	8

global dust, which we implement on a regional basis, as deduced from the data (Kok et al., 2023). Since the elements we consider here are predominantly from dust, we expect, and capture, an approximately 50% increase in these elements near dusty regions (Figure 9). Anthropogenic sources will have increased deposition in some regions as well, although their contribution is much smaller than the dust contributions, and volcanic sources globally (Figure 8b.). The smallest increases are seen in Ca away from the dust source areas, which have large non-dust natural sources; these are not thought to have changed as much since preindustrial times (Figure 9). The exact impact of these changes on deposition are not explored here explicitly, but the monthly mean gridded deposition fields are available for further study.

#### 4. Summary and Conclusions

In this study we compiled available measurements of atmospheric surface concentrations of the dominant elements in dust (Al, Ca, Fe, Ti, and Si), and compared these against new model results focusing on these elements. For the first time, we looked at the non-dust atmospheric sources of Al, Ca, Ti, and Si and modeled their dispersion throughout the atmosphere from their source region to their site of deposition onto the surface. We focus here on the total elemental amounts, not the soluble fraction, because there is more data and knowledge of the total amounts. We acknowledge that soluble fractions can differ substantially from total concentrations, particularly for elements such as Fe, and are more directly relevant for biogeochemical impacts. Therefore, our results should be interpreted as constraints on total atmospheric elemental cycling, rather than bioavailable fluxes. Our goals were to better understand the atmospheric cycling (sources, distribution and deposition) of these elements, estimate where the non-dust sources were important, and consider how the deposition of these elements has changed compared to preindustrial times. We used new mineral estimates from EMIT derived from imaging



**Figure 9.** Increases in elemental deposition since preindustrial times. The ratio of present day to preindustrial annual averaged PM<sub>10</sub> deposition at each grid point, as estimated in the model is shown for (a) Al, (b) Ca, (c) Fe, (d) Si, and (e) Ti.

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spectroscopy measurements from the International Space Station as well as elemental composition studies of the minerals to estimate elemental distributions for these dust-dominated elements. We included both natural and anthropogenic non-dust sources (e.g., Figure 8). Our model-data comparisons suggested that the model was able to capture much of the spatial variability seen in the observations, but there were still significant differences between the model and data that should be resolved in future studies, especially in dust-dominated areas, similar to previous studies (e.g., Rathod et al., 2024). Ca/Al in particular is underpredicting in desert regions, suggesting the under detection of Ca minerals in EMIT (Clark et al., 2024) is playing an important role and should be addressed in the future.

Because these elements originate predominantly from dust, and we followed recent studies that have suggested dust has increased by 55% since preindustrial times (Kok et al., 2023), we estimate a large increase in dust deposition over much of the globe for these elements (Figure 9). Increases were larger for Al, Fe, Si, and Ti, but smaller for Ca, which has more non-dust natural sources (Figure 8b).

For the first time, we estimate the non-dust sources of these elements and show that in many regions, the non-dust sources will dominate. These are sources from both other natural sources (e.g., volcanoes, sea spray, wildfires or primary biogenic particles) or from anthropogenic sources such as fuel combustion. Volcanic sources are estimated here to be the second largest sources, but are poorly represented here: future work should focus on improving the estimates of volcanic sources.

While the role of increased dust-borne Fe deposition to the oceans has been considered in substantial detail (e.g., Kok et al., 2023), changes in atmospheric concentrations and deposition of the other elements (e.g., Al, Ca, Si and Ti) have not yet been considered, although they could be important. In this study, we included a detailed study of both natural and anthropogenic, dust and non-dust sources of these elements which allows for future studies to look in more detail at the possible impacts using gridded monthly mean deposition data for preindustrial and present day.

While our study includes as much in situ concentration data as available, there are regions with little data, inhibiting our ability to verify model estimates against observations (e.g., Figure 1). More data outside North America, Europe, and Japan would enhance our understanding of the distribution of these important elements and their impact on ecosystems.

### Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

### Availability Statement

The model output and observational compilation are Mahowald, Li, Gonçalves Ageitos et al. (2025). The supplemental information also includes the observational compilation and citations.

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