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Can land use intensification in the Mallee, Australia increase the supply of soluble iron to the Southern Ocean?

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The supply of soluble iron through atmospheric dust deposition limits the productivity of the Southern Ocean. In comparison to the Northern Hemisphere, the Southern Hemisphere exhibits low levels of dust activity. However, given their proximity to the Southern Ocean, dust emissions from continental sources in the Southern Hemisphere could have disproportionate impact on ocean productivity. Australia is the largest source of dust in the Southern Hemisphere and aeolian transport of dust has major ecological, economic and health implications. In the Mallee, agriculture is a major driver of dust emissions and dust storms that affect Southeastern Australia. In this study, we assess the dust generating potential of the sediment from the Mallee, analyze the sediment for soluble iron content and determine the likely depositional region of the emitted dust. Our results suggest that the Mallee sediments have comparable dust generating potential to other currently active dust sources in the Southern Hemisphere and the dust-sized fraction is rich in soluble iron. Forward trajectory analyses show that this dust will impact the Tasman Sea and the Australian section of the Southern Ocean. This iron-rich dust could stimulate ocean productivity in future as more areas are reactivated as a result of land-use and droughts.

Satellite observations show that the most active dust sources on Earth are in the Northern Hemisphere^{1,2}, which accounts for 80 to 90% of the global dust emissions³, while the Southern Hemisphere exhibits relatively low dust concentrations. Atmospheric dust is an important source of iron to the high nutrient low chlorophyll (HNLC) waters of the Southern Ocean^{4,5}, although upwelling⁶ and hydrothermal activity⁷ are also considered to be important contributors. Because of the low rates of atmospheric dust deposition in the Southern Hemisphere, the productivity of the Southern Ocean is constrained by the limited supply of soluble iron^{6,8,9}. It has been argued that fluctuations of atmospheric CO₂ concentrations during glacial-interglacial transitions can be explained by changes in supply of iron in dust^{10,11} resulting from changes in dust emissions associated with periods of aridification or revegetation in continental land masses¹².

Presently, the main active dust sources in the Southern Hemisphere include the Lake Eyre Basin in Australia, the Makgadikgadi and Etosha pans in southern Africa, and the Salar de Uyuni region in Bolivia¹³. Land-use practices such as deforestation and overgrazing have potential to activate new dust sources in the Southern Hemisphere^{14,15} with significant impacts to downwind soils and ecosystems. Potential new dust sources that could be activated as an effect of land use have been identified in Patagonia¹⁴, the Kalahari¹⁶, while in Australia the effect of land use on dust emissions has seldom been investigated in the context of its impact on ocean productivity.

Even though the contribution by dust sources in the Southern Hemisphere to the global dust budget is minimal, these sources could have disproportionately large global impact given the proximity to the Southern Ocean⁴. Therefore Australia is an ideal laboratory to study the connection between wind erosion, dust transport and ocean productivity. One of the largest dust sources in the Southern Hemisphere^{13,17}, Australia plays an important role in the supply of dust to the Southern Ocean^{11,18} and adjacent marine and terrestrial systems^{19–22}. Dust storms in Australia generally coincide with periods of drought^{23–25}, and these events can pose serious threats to local economy and human health^{13,26}. For example, it has been estimated that the September 2009 dust storm in New South Wales caused \$299 millions in economic damage²⁷. The impact of soil erosion is often manifested also in a reduction of soil fertility and agricultural productivity²⁸. The increased frequency of dust storms has detrimental effects on human health²⁹; for example dust combined with bushfire smoke led to increased mortality in Sydney³⁰ and widespread respiratory illnesses³¹.


Table 1 | Study sites in the Mallee. The grain size distribution was analyzed for a subsample of the soil collected from the study sites

Site	Latitude (°S)	Longitude (°E)	% clay	% silt	% sand
Dune Crest 1	35.004619	143.248805	1	8	91
Dune Crest 3	35.005234	143.265465	2	8	90
Dune Crest 5	34.993001	143.258671	2	6	92
Interdune 2	35.005002	143.249927	3	14	83
Interdune 4	35.006978	143.270734	4	20	76
Interdune 6	34.994540	143.263006	2	11	87
Salt Pan	35.226154	143.433129	1	15	84
Little Desert National Park	36.527164	142.015167	1	3	96

It is believed that Australia was a significant source of dust during the Last Glacial Maximum³², when increased aridity caused intense deflation of sediments from the continental sources to the Tasman Sea³³. Satellite records suggest that the Lake Eyre Basin is the primary dust source in Australia^{1,2} and episodic dust activity is seen in the Strzelecki Desert^{1,34}. Central Australia and the Mallee within the Murray-Darling Basin are identified as other dust sources³⁵. Although evidence of the extent of wind erosion in the Mallee can be found in land degradation surveys as early as the 1930s³⁶, very little is known about the amount of dust that can be emitted from the Mallee. Numerous studies have investigated the impacts of Australian dust on Southern Ocean biogeochemistry^{5,37–39} or adjacent marine systems^{22,39} however, it is unclear how dust emissions from the Mallee might affect the supply of iron and likely affect the productivity of the Southern Ocean. To this end, this study will assess the dust generating potential of the sediments from the Mallee, analyze the soluble iron content in the dust-sized fraction of the sediments and determine the likely deposition region of the emitted dust.

Other studies^{15,40} have shown that the use of a laboratory dust generator could allow for assessing the potential of sediments to emit dust.

Results

The grain size analysis shows that sediments from the Mallee are sandy with interdunes rich in silt-sized fractions (Table 1). To place these numbers in the broader context of dust emissions potentially supplying iron to the Southern Ocean, we ran the dust generator for sediments from dust sources in Southern Africa, including the southern Kalahari interdunes that could be activated by overgrazing and climate change¹⁵, as well as the currently active dust sources of the Makgadikgadi and Etosha pans (Figure 1a). The dust generating potential for sediments from interdunes and dune crests from the agricultural fields of the Mallee region is greater than the salt pan and the Little Desert (Table 2, Figure 1b). PM_{10} concentrations were on average 74.86 mg m^{-3} for the dune crests and 80.4 mg m^{-3} for interdunes. The PM_{10} concentration of the Little Desert soil is at

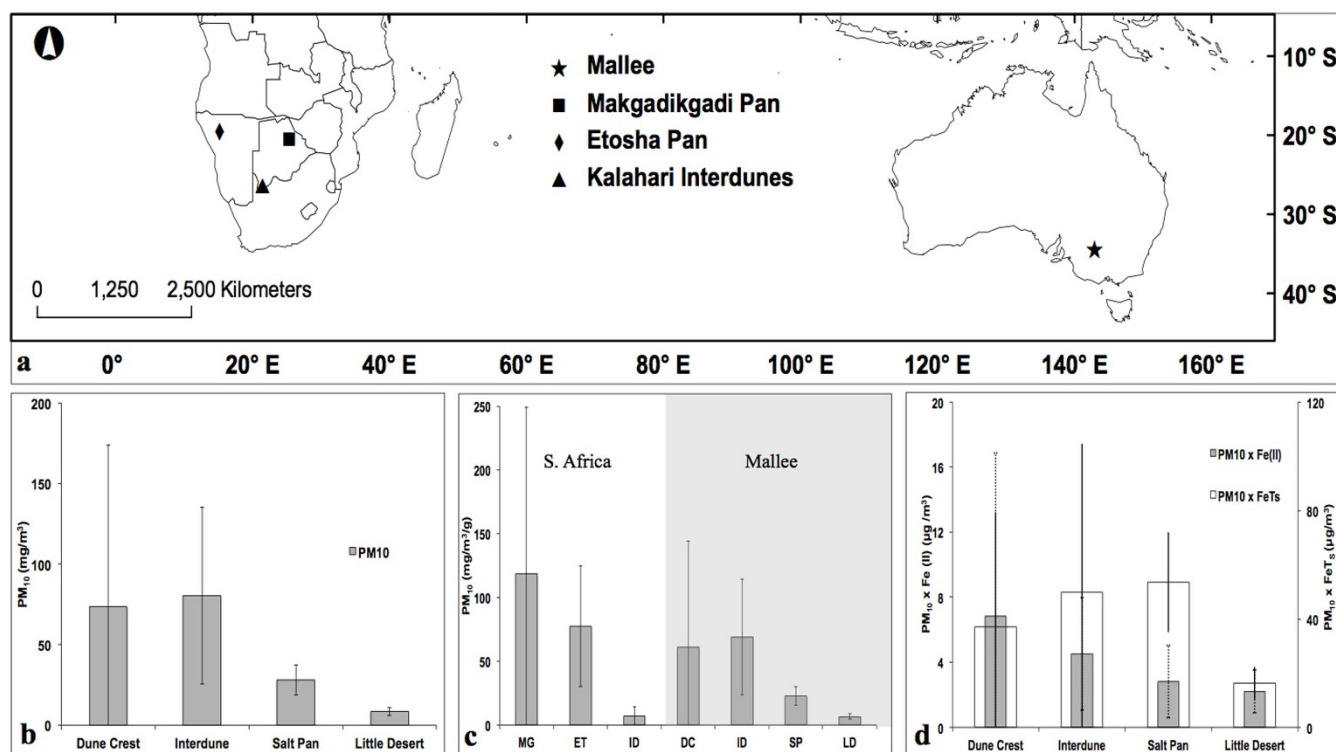


Figure 1 | (a) Map of the Southern Hemisphere dust sources included in this study (b) PM_{10} concentration (mg m^{-3}) from the dust generation experiments with ± 1 standard deviation. PM_{10} values from Little Desert Transect are aggregated. (c) The dust potential per unit mass ($\text{mg m}^{-3} \text{ g}^{-1}$, with ± 1 standard deviation) for the Makgadikgadi Pan (MG), Etosha Pan (ET), southern Kalahari interdunes (ID) and the Mallee dune crests (DC), interdune (ID), salt pan (SP) and little desert (LD) (d) Iron-in-dust potential ($\mu\text{g m}^{-3}$, with ± 1 standard deviation) is calculated by multiplying PM_{10} concentration with the ferrous ion, Fe(II) (grey bars) and with total soluble iron (FeTS) (white bars). (Information on the PM_{10} concentration from Southern African dust sources are available in Supplementary Table S1). This map was created in ArcGIS (ESRI, Redlands, CA).



Table 2 | PM₁₀ values from the dust generator experiment (with ± 1 standard deviation)

Site	PM ₁₀ (mg m ⁻³)
Dune Crest 1	139.67 \pm 172.63
Dune Crest 3	48.65 \pm 5.65
Dune Crest 5	33.26 \pm 19.08
Interdune 2	69.58 \pm 5.39
Interdune 4	128.45 \pm 78.07
Interdune 6	43.17 \pm 10.94
Salt Pan	28.05 \pm 9.26
Little Desert National Park	8.56 \pm 0.55

least one order of magnitude smaller than the interdune and dune sediments (Figure 1b).

The southern Kalahari interdunes have the greatest dust generating potential and thus preferred for this comparison than vegetated dune crests and bare dunes in the region¹⁵. A comparison of dust concentration per unit mass (only for PM₁₀) between the Mallee and the Southern African sediments reveals that the Makgadikgadi Pan sediments have the highest potential (~ 118 mg m⁻³ g⁻¹), followed by the Etosha Pan (~ 77 mg m⁻³ g⁻¹) and the Mallee soils (~ 61 mg m⁻³ g⁻¹ for the dune crests and 69 mg m⁻³ g⁻¹ for interdunes) (Figure 1c). The southern Kalahari interdunes have a potential of about 7 mg m⁻³ g⁻¹. PM₁₀ concentration values for sediments collected from the Makgadikgadi Pan (19–435 mg m⁻³ g⁻¹) and the Mallee dune crests (19–281 mg m⁻³ g⁻¹) have a broad range of variability (Figure 1c).

The soluble iron content is higher in the parent soil for both ferrous ion, Fe(II) and total soluble iron, FeT_s (Table 3). The iron contents of soils from dune crests and interdunes are similar in magnitude for both dust fraction and parent soil. The iron-in-dust potential is a useful measure to assess the potential for soluble iron emissions of different dust sources. It can be expressed as the product of the PM₁₀ concentration to the iron content in the dust fraction. For Fe(II), the iron-in-dust potential is the greatest in soil from the dune crest, followed by interdunes, salt pan and the little desert sediments (Figure 1d). However, for FeT_s, the salt pan and the interdune sediments have comparable iron-in-dust potential (Figure 1d).

The spatial distribution of the terminal location of the 7-day forward trajectories originating at 500 m a.s.l from the Mallee for the period 1999–2009 shows that the recurrent dust transport pathways from the Mallee are to the South Pacific Ocean (Figure 2).

Discussion

Wind erosion is a major cause of loss of soil resources and land degradation in drylands²⁶ and its intensification often results from the expansion of agriculture⁴¹. In Australia, the European colonization led to rapid changes in land use, which contributed to soil erosion and dust emissions⁴². In the case of the Mallee, the driver of degradation was the development of agriculture and overgrazing, which resulted in some of the highest wind erosion rates in

Australia³⁶. Our results show that the potential of the Mallee soils to emit dust is comparable to other currently active dust sources in the Southern Hemisphere (Figure 1c); the deposition of this iron rich dust could possibly alter the productivity and biogeochemical cycling of the South Pacific Ocean.

The percentage of clay and silt sized particles is greater in the Mallee sediments than in the southern Kalahari interdunes (for southern Kalahari grain size data see Bhattachan et al.¹⁶). However, we note that the percentage of clay and silt sized particles in soil is not a robust measure of dust generating potential of a source and other mechanisms such as sandblasting, and breaking up of loose aggregates during erosion could affect the dust generating potential⁴³. Thus, while a laboratory dust generator is useful in quantifying the amount of dust that can be generated by soil from different sources, it cannot simulate the effect of saltation on dust generation. The dust generating potential of the Mallee soil (both dune crests and interdunes) is much greater than the southern Kalahari interdunes (Figure 1c). The Little Desert Transect has the smallest dust potential compared to the interdune and dune sediments however, the dust generating potential of the well-managed farmlands in southern Kalahari is equal to soil found in the protected and managed Little Desert National Park (Figure 1c). It is also interesting to note that the salt pan in the Mallee does not have a high dust generating potential, unlike the salt pans of Southern Africa. This low dust generating potential of the salt pan sediments in the Mallee could be attributed to the lack of ephemeral streams and lunette dunes that supply sediments rich with fine particles, which are prevalent in the Makgadikgadi and Etosha Pans.

The Fe(II) content in the dust fraction between the sediments from the Mallee interdunes and the southern Kalahari interdunes are equal whereas FeT_s is slightly greater in soil in the Mallee⁴⁴. Because soils in Australia are highly weathered, any iron present in such soils is likely to be in hematite and goethite forms, with low amount of ferrihydrite, which in fact is the most labile form of iron oxide²³. Mackie et al.⁴⁵ suggest that ground-based processes such as weathering, and abrasion are credited for low amount of ferrihydrite in dust from Australian sources and any ferrihydrite that is left, it is either transformed to hematite or goethite²³ or lost during saltation because of aeolian abrasion^{46,47}. It is suggested that soluble form of iron is likely to be redistributed to smaller grains during saltation⁴⁵. Our results show that iron present in the Mallee soil is bound in the coarse fraction (Table 3) and the potential of sand grains emitting iron rich fine particles during entrainment could be high. Furthermore, when dust is transported, atmospheric processing especially in clouds is likely to enhance the solubility and therefore bioavailability of iron^{48,49}.

The dust-storm frequency in Australia reaches the highest during austral spring and summer⁵⁰ and the main pathway for dust leaving Australia is over the Tasman Sea and south over the Southern Ocean^{21,38,39}. It has been suggested that the deposition of iron-rich Australian dust in the HNLC waters of the Southern Ocean could impact its productivity⁵. However, in some cases, phytoplankton

Table 3 | Soluble ferrous, Fe(II) and total soluble iron, FeT_s content in mg g⁻¹ of Mallee sediments (with ± 1 standard deviation). D denotes the fine fraction less than 45 μ m and P is the parent soil

Site	Fe (II) _D (mg g ⁻¹)	FeT _{sD} (mg g ⁻¹)	Fe (II) _P (mg g ⁻¹)	FeT _{sP} (mg g ⁻¹)
Dune Crest 1	0.08 \pm 0.03	0.17 \pm 0.17	0.39 \pm 0.23	1.38 \pm 0.10
Dune Crest 3	0.10 \pm 0.05	0.43 \pm 0.64	1.41 \pm 0.39	1.87 \pm 0.01
Dune Crest 5	0.09 \pm 0.06	1.63 \pm 0.65	0.55 \pm 0.72	1.69 \pm 0.22
Interdune 2	0.05 \pm 0.01	0.14 \pm 0.10	0.17 \pm 0.11	1.66 \pm 0.17
Interdune 4	0.07 \pm 0.05	1.01 \pm 0.97	0.08 \pm 0.02	1.88 \pm 0.04
Interdune 6	0.05 \pm 0.06	1.18 \pm 0.72	0.44 \pm 0.21	1.67 \pm 0.08
Salt Pan	0.09 \pm 0.04	1.90 \pm 0.03	0.11 \pm 0.07	1.88 \pm 0.02
Little Desert National Park	0.24 \pm 0.10	1.88 \pm 0.19	1.97 \pm 0.39	2.09 \pm 0.29

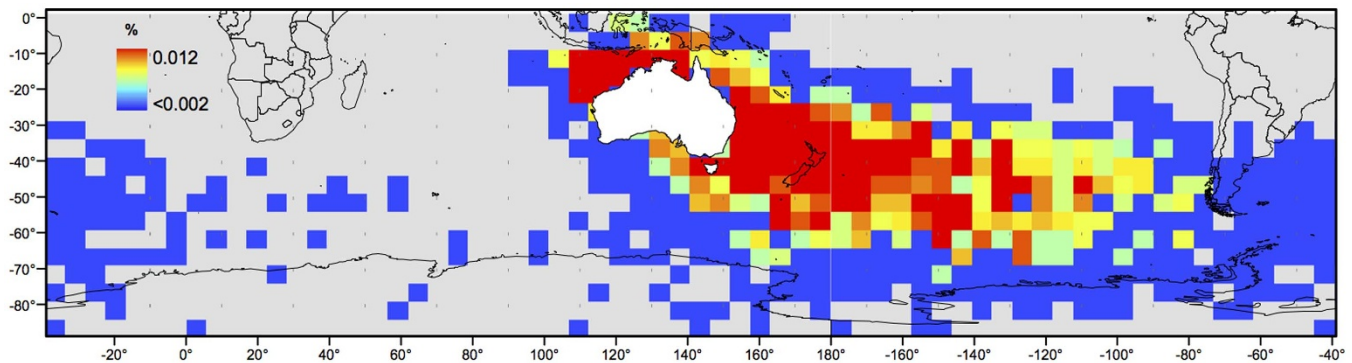


Figure 2 | The spatial distribution of 7-day forward trajectories between 1999–2009 originating from the Mallee initiated at 500 m a.s.l. The percentages are calculated with the total number of terminations in a 5° by 5° grid. This map was created in ArcGIS (ESRI, Redlands, CA).

response was undetected after major dust storms^{18,23} likely because the timing of the event and light availability, which are other important factors controlling ocean productivity. In fact, the deposition of dust is expected to stimulate blooms later in spring or summer when the ocean is more receptive and light is available^{39,51}. Our results from the forward trajectory analysis (Figure 2) show that dust from the Mallee has higher probability of depositing over the Tasman Sea and the Subantarctic Water Ring Province, which is consistent with other studies^{17,21,38,39}. Dust from other sources in Australia, for example, the Lake Eyre Basin (the largest dust source within Australia), however, predominately travels towards the northwest shelf and Great Barrier Reef and not to the Southern Ocean (south of 45° S)⁵¹. Hence, an increase in dust emissions from the Mallee of the Murray-Darling Basin could have significant impacts on the productivity of the Tasman Sea, and the Southern Ocean.

There is some evidence that dunes in Australia can emit dust, particularly after fire induced vegetation loss^{52,53}. Within the Lake Eyre Basin, only 4% of the emissions originated from the actual lakebed whereas dunes accounted for 37% of the plumes⁵². Loss of vegetation cover resulting from increased aridification⁵⁴ and land use (e.g., agriculture, overgrazing) has also been related to the remobilization of stabilized dunefields and enhanced dust emissions in the southern Kalahari¹⁶. Given the potentially strong coupling between Australian land-use practices, dust emissions, and iron delivery to the HNLC waters of the Southern Hemisphere, intensification of agriculture and land degradation in Australia (and the Mallee in particular) could result in an increase in ocean productivity in the South Pacific Ocean.

Methods

Study sites. Triplicated soil samples (about 50 g) were collected from the top 2 cm of soil from six agricultural sites and a salt pan in the Mallee region (Supplementary Figure S1a). These sites are about 2 km away from the banks of the Murray River in an area with a gently rolling topography resulting from relict dunes (Table 1). Additional sampling was performed in the Little Desert National Park (Victoria, Australia) in a duneland that is currently stabilized by grass and woody vegetation (Supplementary Figure S1b). The study site locations for soil samples collected from the southern Kalahari interdunes, and the Makgadikgadi and Etosha Pans are included in supplementary table S1. The grain size distribution of the Mallee soils was measured on a subsample run through a laboratory particle size analyzer (LS 13 320, Beckman Coulter®).

Dust generator. The dust generating potential of the samples collected in the Mallee region of southern Australia and the existing and potential dust sources in Southern Africa was determined using a laboratory dust generator (Custom Products, Big Spring, TX) (Supplementary Figure S1c). Each run was performed with approximately 1 g of soil sample placed in a tube, which rotates around a horizontal pivot axis at the speed of 13 rounds per minute with a 4 s pause between each rotation. A pump draws air from the rotating tube at a rate of 14 L per minute. When the tube is in motion, the sediment falls to the bottom of the tube thereby generating dust. The entrained dust is drawn into the settling chamber where dust concentrations are measured by a dust sensor attached to an aerosol spectrometer (Grimm, Model 1.108). The data units used by the spectrometer are in particle count per liter for particle diameters ranging between 0.3 and 20 µm. The conversion from particle

count per liter to concentration (mg m⁻³) is performed as described in Bhattachan et al.¹⁵. PM₁₀ values (particulate matter less than 10 µm in diameter; expressed in mg m⁻³) are reported in table 2 and table S1.

Soluble iron. The samples were sieved through a 45 µm sieve to separate the dust sized fraction from the parent soil. The dust fraction and parent soil were then analyzed for total soluble iron, FeT_s and soluble ferrous ion, Fe(II). To determine the soluble iron content, 10 ml of 0.5 M HCl were added to 0.25 g of sample and the solution was shaken for an hour, filtered through a nucleopore filter to eliminate particulate matter. An acetate buffer was used to raise the pH to 5.5 and a 0.1 M ferrozine was added to the solution to measure the Fe(II) content on a UV photospectrometer (Shimadzu 4100) at 562 nm⁵⁵. FeTs was measured using the above method after reduction of Fe(II) by adding 0.01 M of hydroxylamine hydrochloride.

Forward trajectory analysis. The 7-day forward trajectories were determined using Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLOT)⁵⁶, originating from a point within the Mallee (35.013256° S, 143.255357° E) for years 1999 to 2009. The model used NCEP/NCAR reanalysis dataset for years 1999–2005 and the GDAS1 data for years 2006–2009. Forward trajectories were run at altitude of 500 a.s.l. every day at 05 UTC.

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Author contributions

A.B. and P.D. designed the study; A.B. performed analysis; and both authors contributed to the interpretation of the results and the writing of the paper.

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