We thank both reviewers and the editor for their constructive comments, which has greatly helped us to improve the paper. Below we include a point-by point response to the referee comments, and describe the corresponding changes we have made to the manuscript.

## V. Etyemezian (Referee #1) comments

This paper examines vertical dust fluxes from a portion of the Oceano Dunes in Coastal California under conditions of windblown transport. It reports on the results from a series of measurements conducted with optical particle counter-style instruments that were spaced vertically to allow for the calculation of dust flux. A second measurement component involved the mineralogical analysis of sand grains to infer the source material of emitted dust. These experiments were conducted simultaneously and in coordination with measurement of sand transport (reported separately). The authors include a discussion of how their results may be applicable to global emissions from sand dunes.

Overall, this was a well written paper on an important topic. I recommend its publication following some revisions. There are some technical areas that ought to be addressed. Additionally, I found that the assertion that the results from this work are relevant for global emissions from sand dunes to be somewhat overstated. I believe this is a stylistics judgement, but have provided comments that the authors may wish to consider.

# We thank the reviewer for his constructive and helpful comments that helped us to further improve the paper. We address these comments below.

The authors rely on the concentration differences between Met One Particle Profiler instruments (model 212) to calculate the vertical flux of dust. If I read correctly, they basically use the two instruments that are mounted closest to the ground. As can be seen from the calibration coefficients in Table S2, these instruments are not exactly the gold standard when it comes to aerosol measurement. They are very useful for the type of field work described, but care should be taken in using the information quantitatively. The inner-instrument comparability changes over time as well as over the concentration range of the measurement. For example, two instruments might have one relationship when the (say) PM 10 concentration is 50 micrograms per cubic meter and another relationship at 300 micrograms per meter cubed. In view of this, it would be important to ascertain that the concentration ranges that were experienced by the instruments during calibration and the ranges experienced during vertical flux measurements were similar. If not, this could be a significant source of bias in the results.

Our figure Fig. S3A in the supplementary material shows that the calibration coefficients are nearly invariant with aerosol concentration; see also Table S2. Nonetheless, we accounted for the variation in the calibration coefficients with aerosol number concentration (see Table S2) and propagated this uncertainty throughout the analysis in the paper. To make this clearer, we added in line 24 of page 4 of the main text that (change underlined) "This procedure yielded a <u>concentration-dependent</u> correction factor with uncertainty for each size bin of each OPC (Table S2), which we propagated throughout our analysis (Fig. S3B)."

Another instrument-related observation is that the results and several of the key findings are related to the fact that the measured particle size distribution (PSD) during wind erosion events appears to become finer with increasing friction velocities. There is the potential that this is a consequence of measurement artifact associated with the changing aspiration efficiency of the inlet with wind speed. Typically, inlets to particulate matter instruments are designed to allow over a range of wind speeds for larger particles (larger than the maximum size of interest) to make the treacherous aerodynamic journey from being in the open air to passing by the actual sensing element, whatever that happens to be. The Met One instrument

almost certainly loses particle collection efficiency at higher wind spends. This loss of efficiency is most pronounced for larger (say >5 microns) particles, giving the appearance that the size distribution if becoming finer at higher wind speeds. The authors should explicitly address/examine this issue and correct any of their results and subsequent conclusions as needed.

We thank the reviewer for this valuable comment, which we now address in the revised version. Unfortunately, the manufacturer of Met One instruments could not share a curve of the dependence of sampling efficiency (i.e., the ratio of particle concentration measured by the sensor to the particle concentration in the ambient air; Von der Weiden et al., 2009) on wind speed and particle size. On the other hand, the study of Von der Weiden et al. (2009) obtained general results on the sampling efficiency as a function of particle size, wind speed, and sensor inlet characteristics. This work supports the reviewer's argument that the efficiency of collection decreases strongly with wind speed for coarse particles, for instance the loss rate can approach 100% for particles larger than 10  $\mu$ m in diameter under strong wind events. Therefore, we have removed the results of our largest bin (Bin 7 with the nominal diameter of 10  $\mu$ m) (see line 31 in page 3, lines 1-5 in page 4, lines 16-23 in page 8, Figs. 3, 4, and 6) and have added a discussion in the limitation section of how the possible decrease of the sampling efficiency for the other bins (notably Bin 6) affects our result that the PSD of emitted dust remains variant with shear velocity in Fig. 4 (see lines 15-19 in page 14).

On a related point, I think the subtraction of the sea salt deposition profile is something of a distraction and perhaps also an artifact of the measurements. However, if the authors choose to retain this portion of the analysis, I would suggest first providing an estimate of the magnitude of sea salt deposition that is inferred and whether this number seems reasonable at all. Second, what mechanism of deposition could account for such deposition rates? Third, is that mechanism shear stress independent as is assumed in the final treatment of the numbers?

This is a good comment. We have added the magnitude of sea salt deposition flux (see line 4 in page 7) and validated against past published study on coastal sea salt deposition (see lines 5-6 in page 7). However, it remains difficult to address the second and third points, because of the complexity of coastal sea salt aerosols. Sea salt emission from coastal ocean increases with shear velocity (O'Dowd and de Leeuw, 2007), and the deposition changes downwind with shear velocity and distance to the shoreline. The sum of those effects depends on a variety of parameters which were not measured in our dataset. On the other hand, based on our available data, we could clearly see the signal of sea salt deposition when saltation is inactive (see lines 24-29 in page 6). Because the assumption of sea salt deposition flux increasing with shear velocity (see Fig. S9), we consider the effect of sea salt deposition to be of second order in our measurements.

On a stylistic point, I can understand that there is a value in expanding the results from the Oceano Dunes to try to understand dust emissions from sand dunes globally. While the authors are careful to caveat their comparisons between the dust emission schemes of (for example) the Sahara and Oceano Dunes, the language in which their work is described blurs out some of the caveats that are stated. For example, in the abstract, it states that "These measurements thus support the hypothesis that considerable emissions of fine dust can be generated by the reactivation of inactive dunes with accumulated clay minerals." Do they, actually?

Elsewhere in the Abstract, these is the statement that "As such, dust emitted from sand sheet, and potentially from other active sands affected by similar dust emission processes, could have potent impacts on climate change, the hydrological cycle, and human health," Sure, this seems quite likely, but it follows

a sentence that starts with "We further fund...", giving the impression that the results of the paper pertain to climate, hydrological, and health impacts of dust from sand dunes.

I have provided examples from the Abstract, but there are several locations in the manuscript where fuzzy sentence transitions amidst caveated statements unintentionally inflate the achievement of the paper. Another example is the series of sentence of page 16 starting on line 4 with "we find that the PSD of dust..." through "... clay coating removal is likely a major emission process for active sands" on line 12. I would urge the authors to check the text for statements that may be misinterpreted as overreaching.

We thank the reviewer for his helpful suggestion. We have revised the abstract and removed "These measurements thus support the hypothesis that considerable emissions of fine dust can be generated by the reactivation of inactive dunes with accumulated clay minerals." from the abstract (see lines 11-20 in page 1). Furthermore, we have systematically rewritten the discussion section, especially section 4.2 about the representativeness of dust emission from Oceano (see lines 12-31 in page 11 and lines 1-14 in page 12) and section 4.3 about the implications for downwind human health, park management, hydrological cycle, and climate (see lines 15-31 in page 12, lines 1-32 in page 13, and lines 1-3 in page 14).

Minor comments: - The legends, axis titles, and figure titles of Fig. S5 are too small to be legible. Suggest making more readable, especially if you elect to retain sea salt deposition corrections.

We have enlarged Fig. S5 in the revised manuscript, detailed in pages 17-18 of the Supplement.

Can you show the size distribution of Bullard et al. Australian laboratory measurements alongside those presented in Figure 3?

We have included Fig. S10 in the revised Supplement that shows the PSD of produced dust by clay-coating removal (Bullard et al., 2004). We chose not to include Bullard et al. (2004)'s results in the main text, because their measured size distributions differ greatly between different times of measurement in their abrasion apparatus (see their Fig. 17) such that no clear conclusions can be drawn from the comparison.

Can you provide a brief explanation (few sentences describing main features of the technique) of how XRPD information is different from SEM-EDS and to what extent either provides bulk versus surface information?

The XRPD method uses data on the crystal structure to retrieve the minerals that make up the bulk sample. On the other hand, SEM-EDS provides information on the coating minerals. We have re-written the final paragraph of section 2 (lines 9-29, page 7) to make this clearer.

Can you provide more detail on how many particles/samples (if applicable) were examined with XRPD and SEM/EDS? I couldn't find details of this work and it seems to be a key point of the paper that the feldspars and clay coatings are important.

We collected 2 soil samples (each ~220 grams) from the tower location and 100 meters upwind, respectively. Four 1-gram samples were examined with XRPD (add on Table 2's caption). Six 1-gram samples were examined with SEM/EDS. We have re-written the final paragraph of section 2 (lines 9-29, page 7) to include these details.

There are likely gray literature reports where the dust emissions were measured at the Oceano Dunes. Naturally, methodology would not be exactly comparable, but it might be worthwhile to ask your ODSVRA contact to provide some of those reports for a gross comparison. Perhaps there is a journal policy on gray literature that precludes this. Thank you for pointing this out. We checked with our Oceano contact (Jack Gillies), who could not point out any relevant gray literature but instead suggested we add the following reference: Gillies, J.A., Etyemezian, V., Nikolich, G., Glick, R., Rowland, P., Pesce, T., Skinner, M., 2017. Effectiveness of an array of porous fences to reduce sand flux: Oceano Dunes, Oceano CA. Journal of Wind Engineering and Industrial Aerodynamics 168, 247–259. https://doi.org/10.1016/j.jweia.2017.06.015. We now refer to this paper as an example of dust emission concentrations measured in the riding area of Oceano Dunes SVRA (lines 28-31 of page 12). We do not know of any published direct comparisons between dust emissions from riding versus non-riding areas at Oceano SVRA.

## Anonymous Referee #2 comments

General comments: This study highlights the significance of sand dunes as an important source of fine dust to the global dust cycle. An in situ field measurement is undertaken at a costal location (Oceanic Dunes) in California to understand the various processes involved in production of aeolian dust from active sands. A suite of instruments were installed in a vertical tower to monitor meteorological and physical parameter to get insight of the physics associated with dust emission from sands. In addition, chemical and mineralogical composition of sand from experimental site were also measured using XRD and SEM-EDS. Based on their result, authors have inferred two major mechanism for fine dust emission, (1) aeolian abrasion of feldspar and (2) removal of clay-mineral coating during saltation, from sands at Oceano site. These inferences are well supported by the previous experiments and literature. The manuscript is well written, easy to read and of great significance for dust research. Thus, I recommend for publication in ACP, however, below I have made few comments which authors may think to consider while revising.

## Thank you for the positive comments. We address your comments carefully as follows.

## Specific comments:

Section 4.2: The discussion on representativeness of Oceano dust emissions can be improved further. As authors have tried to propose "... sand dunes might be globally relevant source of dust." (Page 1 line no. 14-15) and the feldspar abrasion is the major mechanism for such emission. They can include the distribution of feldspar content in active sand dunes and can make an approx. estimation of fine dust emitted by these sources. It is important in order to link its contribution to global dust cycle.

This is a very good suggestion. However, our results are based on only one field site and we cannot separate the effects of emission through clay-coating removal and feldspar abrasion. We thus think that extrapolating our results to estimate dust emissions through feldspar abrasion on a global scale, though clearly desirable, would require substantial future measurements at a range of sand dunes with different properties (including sand size distribution, mineralogy (such as extents of clay-mineral coatings and feldspars), chemical and physical weathering rates, dune type, and palaeoenvironmental history).

Section 4.3: The implication of this study to human health and park management is OK, but stretching it to hydrological cycle and climate is overstatement. This study has not quantified feldspar content being emitted and subsequently carried to higher altitude. So, their role in ice nuclei formation and consequently on the hydrological cycle is highly speculative. The Oceano dust emission might have a local to regional impact. However, the process of dust emission from sand dunes may have global relevance.

We thank the reviewer for these helpful comments. We have re-written section 4.3 about the implications for downwind human health, park management, hydrological cycle, and climate (see lines 15-31 in page 12, lines 1-32 in page 13, and lines 1-3 in page 14) accordingly, and now point out that the impact of Oceano

dust on downwind cloud microphysics remains speculative. We now focused this paragraph more on stressing the need for further research to investigate links of feldspar emission to the hydrological cycle.

Section 4.4: I think authors should have attempted to collect aeolian dust samples on substrate using high volume samplers simultaneously with the other measurements. The enrichment of feldspar in the collected dust samples compared to parent sand had provided robust evidence for the proposed hypothesis of emission of fine dust from feldspar abrasion. This can be included in the limitation section.

Thank you for this suggestion. We now include this as limitation #2 in section 4.4 (see line 8 in page 14).

Table 2: How many sand samples were analyzed to get mineralogical composition? Mention in the table caption.

We collected 4 sand samples (each ~220 grams) in total with 2 samples from the tower location and 2 from 100 meters upwind. For each of the 4 samples, we used 1 gram for XRPD analysis to obtain mineralogical composition of bulk sand grains. We have detailed in Table 2's caption and re-written the final paragraph of section 2 (lines 9-29, page 7) to make this clearer.

## Fine dust emissions from active sands at coastal Oceano Dunes, California

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Abstract. Sand dunes and other active sands generally have a low content of fine grains and, therefore, are not considered to be major dust sources in current climate models. However, recent remote sensing studies have indicated that a surprisingly large fraction of dust storms are generated from regions covered by sand dunes, leading these studies to propose that sand dunes might be globally-relevant sources of dust. To help understand dust emissions from sand dunes and other active sands,

- 15 we present in situ field measurements of dust emission under natural saltation from a coastal sand sheet at Oceano Dunes in California. We find that saltation drives dust emissions from this setting that are on the low end of the range in emissions produced by non-sandy soils for similar wind speed. Laboratory analyses of sand samples suggest that these emissions are produced by aeolian abrasion of feldspars and removal of clay-mineral coatings on sand grain surfaces. We further find that this emitted dust is substantially finer than dust emitted from non-sandy soils, which could enhance its downwind impacts on
- 20 human health, the hydrological cycle, and climate.

#### **1** Introduction

Dust emission by wind-blown (aeolian) processes produces important effects on the Earth system, including modulating the radiation budget (Miller et al., 2006; Kok et al., 2017), modifying cloud microphysics (DeMott et al., 2015) and the hydrological cycle (Ramanathan et al., 2001; Miller et al., 2004), and producing risks to human health (Burnett et al., 2014).

25 Despite these critical impacts, simulations of dust emissions in current climate models have large uncertainties, particularly in predicted rates and spatial patterns of dust emissions (Engelstaedter et al., 2006; Huneeus et al., 2011; Kok et al., 2014b; Evan et al., 2015). These uncertainties arise in part from a lack of understanding of the physics of the emission process, and of the dust emission productivity of different soil types.

Most parameterizations of dust emissions account only for emission through soil aggregate fragmentation, which dominates for non-sandy soils (defined as containing >10% by mass of clay- and silt-sized soil fines; McKee, 1979), and these parameterizations do not account for dust emission through removal of clay-mineral coatings or aeolian abrasion, which might dominate emissions from sandy soils (Shao et al., 1993; Alfaro and Gomes, 2001; Ginoux et al., 2001; Zender et al., 2003; Kok et al., 2014a). For both soil types, dust emission is facilitated by aeolian saltation (the ballistic motion of sand particles) and sandblasting (saltator-driven release of dust) (Shao, 2008). Sandblasting can emit dust in four primary ways: (1)

- 5 fragmentation of aggregates of clay-sized (<2 μm in diameter) and silt-sized (2-63 μm in diameter) fine particles in the soil (Kok, 2011), (2) release of single fine particles trapped between the sand particles (also referred to as "resident fines") (Bullard et al. 2004), (3) rupturing of clay- and oxide-coatings attached to the surfaces of sand grains (Bullard et al., 2004; 2007), and (4) chipping or spalling of sand grains, for instance of sharp corners, which is also known as aeolian abrasion (Kuenen, 1960; Whalley et al., 1987; Jerolmack and Brzinski, 2010; Sweeney et al., 2016). The relative importance of the four physical</p>
- 10 processes depends largely on soil texture and composition: soil aggregate breakage is likely most important for soils with a large fraction of fine (clay- and silt-sized) grains (Shao, 2008; Kok et al., 2012; Swet and Katra, 2016), whereas removal of mineral coatings and aeolian abrasion might be more important for sandy soils (Kuenen, 1960; Bullard et al., 2004; 2007; Jerolmack et al., 2011; Crouvi et al., 2012). Many dust models use preferential source functions to identify emissions in regions with a large content of fine grains held in soil aggregates, which are implicitly used as a proxy for dust emissions through
- 15 aggregate fragmentation (Ginoux et al., 2001; Tegen et al., 2002; Zender et al., 2003). As a consequence, dust models weigh emissions heavily towards non-sandy soils, and sandy soils are not treated as important dust sources in most current models (Shao et al., 1993; Ginoux et al., 2001; Tegen et al., 2002; Zender et al., 2003; Ito and Kok, 2017).

Although current models do not treat active sands (i.e., sands with active saltation transport; Swet et al., 2019) as major dust sources, recent remote sensing observations found a higher correlation between dust emission events and wind strength over

- 20 dune-covered surfaces than over many other geomorphic units of the Sahara Desert, the Chihuahuan Desert, the Lake Eyre Basin, and the Taklamakan Desert (Baddock et al., 2011; Bullard et al., 2011; Crouvi et al., 2012). Specifically, Crouvi et al. (2012) found that more than 40% of Saharan dust storms originated from sand dunes, and that sand dunes had a higher land erodibility (regression of number of dust storms against surface wind) than other soil types. Based on these observations, Crouvi et al. (2012) proposed aeolian abrasion on active sand dunes as an important dust emission mechanism. Although
- 25 remote sensing observations are informative for regional dust emission patterns (Prospero et al., 2002; Crouvi et al., 2012), uncertainties in back-tracking satellite images to locate exact dust sources, combined with the lack of information on dust concentration or composition, small-scale land-surface dynamics, and magnitude of wind stress (Bullard et al., 2011), impede resolving the productivity of active sands.

Considering these limitations of satellite observations, field measurements of dust emission from active sands are needed to inform whether active sands are indeed important contributors to the global dust cycle and, if so, what the physics of the emission process and the dust emission productivity of active sands are. Unfortunately, there are few measurements of dust emissions from active sands, especially under natural field conditions (Bullard et al., 2004; Sweeney et al., 2016; Swet et al., 2019). To improve our understanding of dust emission from active sands, we present the first (to our knowledge) in situ field measurements of dust emission under natural aeolian saltation from active sands at the coastal Oceano Dunes in California. Section 2 details the field campaign setup, in situ data processing methods, and laboratory techniques used to analyse sand samples. Section 3 presents results of the vertical dust mass flux, the particle size distribution of emitted dust, the size-resolved

5 sandblasting efficiencies, and the properties of sand samples. Section 4 discusses dust emission processes at the study site and their representativeness for dust emissions from other active sands, and the implications of our results for downwind human health, the hydrological cycle, and climate. Conclusions follow in Section 5.

#### 2 Methods

We conducted a field campaign from May 15th to June 7th, 2015 on the southern edge of the Oceano Dunes State Vehicular

- 10 Recreation Area (35.03°N,120.63°W) where off-highway motor vehicles were prohibited (Oceano Dunes SVRA map: <a href="http://ohv.parks.ca.gov/?page\_id=1208">http://ohv.parks.ca.gov/?page\_id=1208</a>). At the time of measurements, the site was not downwind of any vehicle activity and was completely nonvegetated and topographically mostly flat. This active coastal sand sheet is followed inland by low transverse dunes. These dunes are part of the broader Late Quaternary Guadalupe-Nipomo Dune Complex, which extends north-south along roughly 25 kilometers of California's Central Coast and is shaped by strong onshore (westerly) sea breezes
- 15 transporting sand primarily derived from fluvial deposits of the Santa Maria River estuary at the south end of the dune field (Cooper, 1967; Orme, 1992; Pye and Tsoar, 2009). Currently, active aeolian transport occurs only within 1-2 kilometers of the beach, but inactive, vegetation-stabilized transverse dune surfaces extend up to 18 kilometers inland.
- We erected a 10-meter tower (Fig. 1) at a distance of ~650 meters from the shoreline to measure wind speed and direction, sand flux, and dust concentration from May 15<sup>th</sup> to June 4<sup>th</sup>, 2015. We mounted six Campbell Scientific CSAT3 sonic anemometers (Liu et al., 2001) on the tower, of which we used the three-dimensional winds measured by the lowest sonic anemometer (about 0.5 meters above the surface) to determine shear stress and shear velocity (details in Martin and Kok, 2017). In addition, we obtained the vertical profile of horizontal sand saltation flux q(z) from nine Wenglor YH03PCT8 electric laser gate sensors (Barchyn et al., 2014), mounted at heights *z* spanning from 0.02-0.47 meter above the surface. We converted the measured particle count to sand flux by calibrating to eight concurrently sampling Big Spring Number Eight
- 25 (BSNE) samplers (Fryrear, 1986; Goossens et al., 2000). We then obtained the total sand flux by integrating the flux through the depth of the saltation layer (see Martin and Kok, 2017; Martin et al., 2018). Furthermore, we obtained the dust concentration profile using six identical optical particle counters (OPCs) (the 212 ambient particulate profiler, manufactured by Met One Instruments, Inc.) of which we mounted four on the tower at four different heights at any given time within 0.74-6.44 meter above the surface (location details in Table S1). Each OPC measured size-resolved aerosol concentrations using seven size
- 30 bins with equivalent light scattering diameter ranges within 0.49-10  $\mu$ m, calibrated using polystyrene latex spheres (PSLs) (Table 1, column 1). Of the seven size bins, we only used the smallest six bins. Past studies have found that the sampling

efficiency (i.e., the ratio of particle concentration measured by the sensor to the particle concentration in the ambient air; Von der Weiden et al., 2009) can decrease strongly with wind speed for coarse particles; for instance, the loss rate can approach 100% for particles larger than 10 µm in diameter under strong wind events (Von der Weiden et al., 2009). As the dependence of sampling efficiency on wind speed and particle size was not provided by the instrument's manufacturer, we did not use the

5 largest size bin (bin 7) and only use the smallest six bins (with PSL diameter ranging 0.49-7 μm, see Table 1).

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The optical sizing of aerosols is sensitive to differences in the refractive index between the measured aerosols and the particles used to calibrate the instrument (Hinds, 1999). The manufacturer calibrated the OPCs against PSLs, following the international standard ISO 21501-1:2009 (ISO, 2009). We corrected the manufacturer-provided bin size limits to dust size limits using Lorenz-Mie theory (Bohren and Huffman, 1983; Mätzler, 2002; Rosenberg et al., 2012), thereby approximating dust as spherical particles. Specifically, we calculated the light scattered by PSLs (refractive index: m = 1.59 - 0i) at the instrument's laser wavelength (589 nm) and scattering angle range (90° ± 60°; information provided by Met One Engineering Department), using the manufacturer-provided size boundaries of each of the six OPC bins. For each bin, we then determined the size range of dust particles that would produce an equal range of scattered light, using globally-representative dust refractive indices (real part  $n = 1.53 \pm 0.03$  and imaginary part  $k = -10^{-2.5\pm0.3}$ , after Kok et al., 2017). This operation yielded the corrected bin size boundaries for dust and their uncertainties (Table 1, column 2, Fig. S4, and Section 2 in the Supplement).

Using the instrumentation and procedure mentioned above, we obtained shear stress, total sand flux, and size-resolved dust concentration, which we averaged over 30-minute periods. We chose a 30-minute averaging time interval because this interval is shorter than the typical time scale of a mesoscale weather system but sufficient to capture most of the turbulent energy spectrum (Stull, 1988; Sterk et al., 1998). Furthermore, we intercalibrated the six OPCs to reduce systematic errors in the

- 20 measured dust concentration and to quantify the instrument uncertainty. To obtain measurements for this intercalibration, we mounted five OPCs (the sixth one malfunctioned after May 25<sup>th</sup>, 2015) at the same height and in a line perpendicular to the wind for three days (June 5<sup>th</sup>, 6<sup>th</sup>, and 7<sup>th</sup>, 2015) immediately following the field campaign (Fig. S2). Specifically, for each of the six size bins, we applied linear-least squares regression on the aerosol concentration of each of the five OPCs against the mean of the five OPCs (Fig. S3A). This procedure yielded a concentration-dependent correction factor with uncertainty for
- 25 each size bin of each OPC (Table S2), which we propagated throughout our analysis (Fig. S3B). For all of our subsequent analysis, we did not use dust concentrations measured by the malfunctioning OPC, because they did not satisfy our data-quality control criteria (for more details, see Section 1 in the Supplement).

We used the calibrated dust concentration to obtain the size-resolved mass flux of emitted dust using the gradient method, which is analogous to the methods for determining vertical scalar fluxes from turbulent and molecular diffusion in the

30 atmospheric surface layer (Gillette et al., 1972; Shao, 2008). The gradient method assumes constant dust flux within the surface layer, neutral atmospheric stability (see Fig. S2 of Martin and Kok, 2017), and negligible dust deposition from upwind sources, and is expressed as

$$F_{d,i} = -K_d \frac{\partial c_i}{\partial z},\tag{1}$$

$$K_d = \eta \kappa u_* z , \tag{2}$$

where  $F_{d,i}$  is the vertical mass flux (kg/m<sup>2</sup>/s) of the *i*<sup>th</sup> size class of the six bins,  $c_i$  denotes the mass concentration (kg/m<sup>3</sup>) of the *i*<sup>th</sup> size class at height *z* (m),  $K_d$  is the turbulent diffusivity (m<sup>2</sup>/s) obtained by the mixing length theory (Stull, 1988),  $\eta$ 

5 is the ratio between the turbulent diffusivity of a passive tracer and that of momentum, which we take as unity based on previous studies (Gillette et al., 1972; Stull, 1988),  $\kappa$  is the von Kármán constant, which we take as 0.387 (Andreas et al., 2006), and  $u_*$  is the shear velocity (m/s). Combining Eqs. (1) and (2), and integrating from a reference height  $z_r$  (m), we obtain

$$c_i(z) = c_i(z_r) - \frac{F_{d,i}}{\kappa u_* \eta} ln\left(\frac{z}{z_r}\right),\tag{3}$$

- 10 where  $c_i(z_r)$  is the reference concentration of the *i*<sup>th</sup> size class at  $z_r$ . For calculation purposes, we set  $z_r$  as the height of the "D1" OPC (Fig. 1 and Table S1, column 2). Applying Eq. (3) to each of the six bin classes yields the size-resolved vertical dust mass flux  $F_{d,i}$  for each bin. We then obtained the bulk vertical mass flux as the sum of the size-resolved fluxes  $F_d = \sum_{i=1}^{6} F_{d,i}$ . Note that for each bin we only used measurements that showed a negative gradient (concentration decreases with height) and thus a positive dust flux, as discussed further below.
- 15 We used the size-resolved vertical dust flux obtained above to calculate the particle size distribution (PSD) of emitted dust and the sandblasting efficiency, which together give insights into the physical processes governing dust emission from active sands (Kok et al., 2014b; Mahowald et al., 2014). We computed the normalized volume PSD of dust at emission as

$$\frac{dV_i}{d\ln D_i} = \frac{F_{d,i}}{F_d \cdot \ln(D_{i+1}/D_i)},\tag{4}$$

where  $D_i$  (µm) and  $D_{i+1}$  (µm) are the lower and upper boundary geometric diameter sizes, respectively, of the *i*<sup>th</sup> size class 20 (Table 1, column 2). The integral of  $\frac{dV_i}{dlnD_i}$  over particle size thus yields unity. For each size bin we then obtained the sandblasting efficiency, which is the vertical dust flux produced by a unit horizontal sand saltation flux (Marticorena and Bergametti, 1995),

$$\alpha_i = \frac{F_{d,i}}{Q},\tag{5}$$

where Q (kg/m/s) is the total horizontal sand flux integrated over all sand grain sizes (see Martin and Kok, 2017). We then obtained the bulk sandblasting efficiency  $\alpha$  (m<sup>-1</sup>) by summing over the six  $\alpha_i$  (m<sup>-1</sup>),  $\alpha = \sum_{i=1}^{6} \alpha_i$ . We used the size-resolved vertical mass flux calculated above to obtain the vertical flux for particulate matter with geometric diameter  $D_g \leq 10 \ \mu m \ (PM_{10,g})$  and with aerodynamic diameter  $D_a \leq 2.5 \ \mu m \ (PM_{2.5,a})$ . Dust in atmospheric circulation models is usually represented in terms of geometric diameter (Mahowald et al., 2014), and the PM<sub>10,g</sub> size range is considered most relevant to dust impacts on weather and climate (Kok et al., 2017). In contrast, aerodynamic diameter is more relevant to

5 aerosol impacts on human health, which corresponds with the  $PM_{2.5,a}$  concentration (Burnett et al., 2014). To obtain the  $PM_{10,g}$  flux, we summed the mass flux of the smallest six size bins and part of the seventh size bin that is within the  $PM_{10,g}$  size range (see Table 1, column 2), for which we integrated over the sub-bin size distribution obtained by linear-least squares regression on the PSD of the emitted dust of the sixth and seventh size bin. To obtain the  $PM_{2.5,a}$  flux, we first converted the geometric diameter bin sizes to aerodynamic diameter bin sizes through (Hinds, 1999)

$$10 \quad D_g = \sqrt{\frac{\chi \rho_0}{\rho_P}} \ D_a \ , \tag{6}$$

where  $D_a$  and  $D_g$  are aerodynamic and geometric diameter, respectively,  $\rho_0 = 1000 \text{ kg/m}^3$  is the density of water,  $\rho_P \approx (2.5 \pm 0.2) \times 10^3 \text{ kg/m}^3$  is the typical density of dust aerosols (Kok et al., 2017), and  $\chi$  is the dynamic shape factor, which is defined as the ratio of the drag force experienced by the irregular particle to the drag force experienced by a spherical particle with diameter  $D_g$  (Hinds, 1999). We used  $\chi \approx 1.4 \pm 0.1$  (Kok et al., 2014a), which yielded  $D_g \approx (0.75 \pm 0.04)D_a$  (values of geometric and aerodynamic bin diameters are listed in Table 1). Second, we summed the mass flux of the smallest two size bins and part of the third size bin that is within the PM<sub>2.5,a</sub> size range (see Table 1, column 3), for which we integrated over the sub-bin size distribution obtained by linear-least squares regression on the PSD of the emitted dust of the second and third size bin.

15

We found that aerosol concentration, vertical mass flux, and the PSD calculated with the procedure above were affected by 20 both dust emission and sea-salt aerosol deposition. Specifically, we found deviations in the measured aerosol concentration profiles (Fig. S5) from the logarithmic profile expected to occur from an active dust emission source (Stull, 1988; Kind, 1992; Gillies and Berkofsky, 2004), a result that we inferred as the influence of sea-salt aerosol. Because we measured dust concentrations ~650 meters from the shoreline, we expect increasing sea-salt aerosol concentration with height due to the upwind deposition of near-surface sea-salt aerosol (Dueker et al., 2011; Liang et al., 2016). We generally observed an

- increasing concentration with height for the lowest two or three OPCs when saltation was inactive (horizontal saltation flux Q = 0), consistent with sea-salt aerosol deposition, but found a decrease in concentration with height when saltation was active (Q > 0), consistent with dust emission (Fig. S5). Furthermore, the measured PSD was coarser when saltation was inactive than when it was active (Fig. S6). This observation is consistent with sea-salt aerosol being coarser than dust aerosol (O'Dowd and de Leeuw, 2007), and dominating when dust emission is not occurring. We thus mitigated the problem of the
- 30 influence of sea-salt aerosol on our results by using only the lowest two sensors (D1 and D2 in Fig. 1), which were most affected by dust emission and least affected by the upwind sea-salt aerosol emission. Indeed, using only the lowest two sensors

caused the aerosol flux to be small and negative (deposition) when saltation was inactive, and large and positive (emission) when saltation was active (Fig. S7). Because using the lowest two sensors did not eliminate the deposition flux of sea-salt aerosol from our results, we in addition subtracted mass flux measured by D1 and D2 when saltation was inactive from the flux by D1 and D2 when saltation was active. We found the sea-salt deposition flux at Oceano was of the order of  $0.1 \,\mu g/m^2/s$ 

5 (Fig. S8), which was roughly an order of magnitude smaller than measurements 50 meters away from the shoreline in coastal Maine, USA (Dueker et al., 2011). Due to a lack of precise sea-salt deposition data in our measurements, we assumed the seasalt aerosol deposition flux to be invariant to shear velocity (Fig. S8A). However, accounting for the possible dependence of sea-salt deposition flux on shear velocity does not qualitatively affect the PSD of dust at emission (Fig. S9).

In order to characterize the sand grains at the experimental site, we collected four sand samples (each ~220 grams) on October

- 10 14<sup>th</sup>, 2016 from the upper 2 centimeters of the surface, with two samples at the tower location and two at 100 meters upwind. We analyzed the properties of each sand sample using a series of physical and chemical techniques. First, we analyzed the particle size distribution (PSD) using the laser diffraction technique with the ANALYSETTE 22 MicroTec Plus, Fritsch (Katra and Yizhaq, 2017), which measured particles within 0.08-2000 µm in optical diameter. We calculated the PSD of soil samples with a Fraunhofer diffraction model with a size resolution of 1 µm using MasControl software (Katra and Yizhaq, 2017). Note
- 15 that we did not convert this soil PSD in terms of optical diameter into geometric or aerodynamic diameter due to a lack of information in particle shape, refractive index, scattering angle range, and laser wavelength. Second, we analyzed the mineralogical composition of the sand grains using the X-ray powder diffraction (XRPD) method (Klute, 1986). The XRPD method allows the crystal structure data to retrieve the minerals of a bulk sample. Each analysed sample of 1 gram contain hundreds of grains at the sand size of the Oceano site. Specifically, we used the PANalytical Empyrean Powder Diffractometer
- 20 equipped with position sensitive detector X'Celerator (Philips 1050/70). Data were collected in the  $\theta/2\theta$  geometry using Cu K<sub> $\alpha$ </sub> radiation ( $\lambda = 1.54178$ Å) at 40 kV and 30 mA. Scans were run over ~15-minute intervals in a 2 $\theta$  range of 4-60° with a step equal to ~0.033° (Sommariva et al., 2014). The Reference Intensity Ratio (RIR) method was used to determine the concentrations of the crystalline components. The integral intensities of main peaks were taken for computation (Gualtieri, 1996; 2000). Third, we performed a qualitative examination of the grain surfaces using Scanning Electron Microscopy (SEM)
- 25 (Quanta 200, FEI) and the Energy Dispersive X-ray Spectroscopy (EDS) chemical analysis technique integrated within SEM. The SEM-EDS stimulates each sample by electrons and detects the spectrum of outgoing photons to retrieve elemental analysis of the grain surfaces, which provided us information on mineralogy of grain coatings. In our case, each sample replica was composed of some dozens of particles, including sand-sized and dust-sized particles. It additionally provided images at extremely high magnification (6x to 1,000,000x) that enables a close analysis of the smallest dust particles (<2 μm).</p>

#### **3** Results

15

We find that dust emitted from the sand sheet at Oceano differs from dust emitted from non-sandy soils in two key ways. First, the vertical  $PM_{10,g}$  and  $PM_{2.5,a}$  dust fluxes at Oceano are both smaller than those fluxes from most non-sandy soils at the same shear velocity (Fig. 2). The vertical  $PM_{10,g}$  and  $PM_{2.5,a}$  fluxes at Oceano range from 1 to 100 µg/m<sup>2</sup>/s and 0.1 to 30 µg/m<sup>2</sup>/s,

- 5 respectively. They both increase non-linearly with increasing shear velocity in the measured range of 0.29-0.43 m/s. For similar shear velocities, the  $PM_{10,g}$  and  $PM_{2.5,a}$  dust fluxes from most non-sandy soils exceed those at Oceano, differing by a factor of ~1-100 and ~0.1-10, respectively. The second key difference is that dust emitted from the Oceano site is substantially finer than size-resolved dust emitted from non-sandy soils under natural saltation (Fig. 3A), and it is also significantly finer than dust measured in situ over North Africa (Fig. 3B), which likely accounts for a majority of the world's dust emission (Prospero
- 10 et al., 2002; Engelstaedter et al., 2006).

Our measurement of the bulk sandblasting efficiency provides further insight into the differences between dust emissions from the Oceano sand sheet and from non-sandy soils. First, the bulk sandblasting efficiency at Oceano is around  $10^{-6}$  m<sup>-1</sup> (Fig. 5), which is substantially smaller than the range within  $10^{-5}$  to  $10^{-2}$  m<sup>-1</sup> typical for non-sandy soils (Kok et al., 2012). Second, the bulk sandblasting efficiency increases non-linearly as a power law in shear velocity (Fig. 5), a result consistent with some previous studies (Shao et al., 1993; Marticorena and Bergametti, 1995; Kok et al., 2012).

We find that the dependence of sandblasting efficiency on shear velocity changes with dust size (Fig. 6), which explains the shift of the PSD of emitted dust to finer size with increasing shear velocity (Fig. 4). Specifically, the power law exponents of the dependence of sandblasting efficiency on shear velocity are large and positive for the smallest five bins, whereas the exponent substantially drops to a negative value for bin 6 (Fig. 6). This explains the fining of the PSD of emitted dust with increasing shear velocity, which is almost entirely due to a decreased contribution of bin 6 (Fig. 4). We consider it likely that

<sup>20</sup> increasing shear velocity, which is almost entirely due to a decreased contribution of bin 6 (Fig. 4). We consider it likely that this decreased contribution from bin 6 (with  $D_g$  ranging 6.21 to 9.14 µm; see Table 1) with increasing shear velocity is caused by a decrease in sampling efficiency of coarse particles with wind speed (Von der Weiden et al., 2009). Therefore, it is possible that the PSD of emitted dust remains approximately invariant with shear velocity.

Our analyses of sand grains sampled from the field site show that the Oceano sand is characterized by a unimodal size distribution (mode at 461 µm and median at 491 µm) with 0.95% (by mass) of loose clay- and silt-sized fine particles (<63 µm) between the sand grains and 0.41% of particulate matter smaller than 10 µm (Fig. 7). The sand consists of a mixture of quartz (51% by mass), feldspars (K-rich feldspar 23% and plagioclase 23%), and clay minerals (3%) analyzed by the XRPD technique (Table 2). We further find the presence of mineral coatings on the top of quartz sand grains through the SEM-EDS technique (Fig. 8D and 8E). Note that the mass content of clay minerals detected in the XRPD analysis can be in the form of loose fine dust in the pore spaces between the sand grains and/or clay-mineral coatings attached to the sand grains. In addition,

we find that the surface of the feldspars appears more abraded than the solid surface of the quartz sand grains (compare location

"B" and location "C" in Fig. 8A), which are representative of a large number of SEM-EDS observations made for these samples.

#### 4 Discussion

We reported in situ field measurements of natural dust emission from an undisturbed coastal sand sheet at Oceano Dunes in **California** (Figs. 1 and S1). We found that dust emission from these active sands differs in several key ways from dust emission from non-sandy soils, namely in the magnitude of vertical PM<sub>10,g</sub> and PM<sub>2.5,a</sub> dust fluxes (Fig. 2), the particle size distribution (PSD) of dust at emission (Fig. 3), and the magnitude and the shear velocity dependence of sandblasting efficiency (Fig. 5). Furthermore, we found that sand grains at the study site are coarse with a mode larger than 460 μm (Fig. 7), that many sand grains contain clay-mineral coatings, and that about half of the sand grains are feldspars (K-rich feldspar and plagioclase)

10 (Table 2). These results provide insights into several fundamental questions: (1) what physical processes drive dust emission from active Oceano sands? (2) how representative are dust emissions from the sand sheet at Oceano of active sands elsewhere? and (3) what are the implications of dust emission from Oceano for downwind human health, park management, hydrological cycle, and climate? After addressing these questions, we end the discussion section with several limitations of our methodology.

#### 15 4.1 Insights into processes producing dust from active sands at Oceano Dunes State Park

Our results indicate that aeolian abrasion of feldspars is one possible dust emission process at Oceano. XRPD analysis confirms the existence of feldspars (~46% by mass) (Table 2). Although the content of feldspars is of the same magnitude as that of quartz (~51% by mass) (Table 2), the surfaces of the feldspars commonly appear more abraded than the surfaces of the quartz sand grains (Fig. 8A), suggesting a higher potential to generate dust through aeolian abrasion of feldspars than of quartz grains.

- 20 We list four supporting findings from other experimental studies. First, both laboratory (Dutta et al., 1993; Jari, 1995) and wind tunnel (Kuenen, 1960; 1969) experiments have found that feldspars are more fragile than quartz grains to aeolian abrasion, possibly owing to the cleavage structures of feldspars (Kuenen, 1969) and the greater propensity of feldspars to weather than quartz (Nesbitt et al., 1997). Second, the feldspar content of dust is higher than that of the parent top soils in dust-producing regions, including Northern Ghana (Tiessen et al., 1991), China (Feng et al., 2008), and the UK (Moreno et al.,
- 25 2003), implying the preferential generation of dust from feldspars. Third, laboratory experiments have found that aeolian abrasion of feldspars can generate a larger fraction of the  $PM_{10}$  and the  $PM_{2.5}$  size ranges than generated from abrasion of quartz (Domingo et al., 2010). Indeed, laboratory experiments on washed and clean quartz grains (for which grain surface minerals have been removed) (Whalley et al., 1987; Bullard et al., 2007) and on freshly crushed quartz grains (Wright et al., 1998; Wright, 2001) imply that the dust produced by aeolian abrasion of quartz grains has a small portion (less than 1% by
- 30 mass) within  $PM_{10}$  size range. Fourth, Pye and Sperling (1983) found that when coastal dune sands are exposed to salt solutions under desert diurnal temperature and humidity cycles, feldspars are more susceptible to breakage into fine grains than quartz.

Because the Oceano measurement site is exposed to high levels of sea-salt aerosols deposition (see Methods), this finding further supports our hypothesis that aeolian abrasion of feldspars is one possible dust emission process at Oceano.

Our results further suggest that the removal of clay-mineral coatings by saltator impact is another possible contributor to dust emissions at Oceano, which is supported by the following two findings. First, XRPD analysis found the content of clay minerals

- 5 to be around 3% by mass (Table 2) for Oceano sand samples, although the fraction of this contributed by clay-mineral coatings is uncertain because XRPD analysis cannot distinguish between clay-mineral coatings and loose clay-sized fines trapped in the pore spaces between sand grains (Swet et al., 2019). However, the PSD of dust emitted from the Oceano site is generally consistent with the PSD of emitted dust of laboratory experiments on sand samples taken from Australian active sand dunes (Fig. S10) (Bullard et al., 2004; 2007). Specifically, Bullard and co-workers simulated aeolian abrasion within an air chamber
- 10 on the Australian sand grains with the content of clay-mineral coatings as  $\sim 1\% 2\%$  by mass. They concluded that the removal of clay-mineral coatings was likely the main process for PM<sub>10</sub> dust emissions, implying that the removal of clay-mineral coatings is a key emission process at Oceano as well. Second, a companion paper reports results of wind tunnel experiments on sand grains collected from our Oceano field site, as well as from two active desert dune fields in Israel (Swet et al., 2019). They reported that all three sands produced substantial dust emissions, and their measured sandblasting efficiency of Oceano
- 15 sands was consistent with our field measurements. After Swet and co-workers washed the sand grains from the two Israeli dune fields to remove most of the loose resident fines, leaving the clay-mineral coatings largely unchanged, they found that the washed sand grains still produced  $PM_{10}$  dust emissions that were comparable to those before the sand was washed. Although Swet et al. (2019) did not perform this last experiment on Oceano sand, these results support the interpretation that removal of clay-mineral coatings is a key process driving dust emissions from active sands, including at Oceano.
- Our results imply that sandblasting of resident fines by saltators is a third possible dust emission process at Oceano. The resident fines exist as single particles trapped in between the sand grains, as was observed when we washed sand samples in the lab. Although XRPD analysis cannot distinguish between clay-mineral coatings and clay-sized resident fines in the pore spaces between sand grains (Swet et al., 2019), the content of these resident fines is less than 3% by mass (Table 2). Because we observed a net dust emission flux only when saltation was occurring (Figs. S5 and S7), it is unlikely that the direct aerodynamic entrainment of resident fines (Klose and Shao, 2012) contributed substantially to dust emissions at Oceano. The occurrence of dust emission by removal of resident fines is supported by the measurements of Swet et al. (2019), which reported that unwashed sands produced more dust than washed ones.

Our results provide two insights into the energetics of dust emission from Oceano. First, the lower bulk sandblasting efficiency at Oceano, compared to that of non-sandy soils, supports the common hypothesis that sandblasting efficiency increases sharply

30 with clay-sized grain content (Marticorena and Bergametti, 1995; Kok et al., 2014a). Second, we find that the bulk sandblasting efficiency of Oceano sand increases as a power law with shear velocity (Fig. 5). Because such enhancement of sandblasting efficiency occurs despite invariance of mean saltator velocity with shear velocity (Martin and Kok, 2017), such changes in

sandblasting efficiency must be driven instead by changes in the probability distribution of the saltator impact energies. The recent dust-emission model of Kok et al. (2014a) predicts a power law increase of the sandblasting efficiency with shear velocity for soils for which the typical saltator impact energy is substantially less than the threshold impact energy needed to overcome particle bonds. This power law increase occurs because, for such erosion-resistant soils, only particularly energetic

- 5 saltator impacts are capable of emitting dust, and the fraction of saltators impacts that are particularly energetic increases nonlinearly with wind shear velocity (see Fig. 16 in Kok et al., 2012; Fig.1 in Kok et al., 2014a). As such, our results both tentatively support the Kok et al. (2014a) dust-emission model and suggest that dust emission from Oceano sands are predominantly produced by saltators with an impact energy much greater than the mean impact energy (i.e., with impact energy in the high-energy tail of the saltator-impact-energy distribution). This dependence of dust emission on the very energetic
- 10 saltator impacts likely occurs because both removal of clay-mineral coatings and aeolian abrasion of feldspars are energetic processes, and thus require particularly energetic saltator impacts.

#### 4.2 Insights into representativeness of Oceano dust emissions for other active sands

It is unclear how representative our measurements on the active coastal sand sheet at Oceano are of dust emission from other active sands, including active sand dunes, in particular in relation to the sand size distribution, the content of feldspars, and the extensiveness of clay-mineral coatings.

15

Our results indicate that sand at our study site, with a mode around 461 µm (Fig. 7) is coarser than sand of most active sand dunes (Bullard et al., 2004; Pye and Tsoar, 2009; Webb et al., 2013; Swet et al., 2019). Because the threshold shear velocity required to sustain saltation increases with the grain particle size (Shao, 2008; Kok et al., 2012), the value of 0.28 m/s we found at Oceano (Martin and Kok, 2017; 2018) is likely somewhat higher than that at typical active sand dunes, as also

20 confirmed in the laboratory measurements of Swet et al. (2019). As such, the horizontal saltation flux at Oceano could be somewhat lower compared with typical active sand dunes.

Our results further indicate that Oceano sand differs from sand of typical active sand dunes in its mineralogy, specifically in the content of feldspars. Oceano sands contain as much as ~46% feldspar by mass (Table 2). This content is approximately twice the proportion of feldspar found in coastal dunes along Monterey Bay to the north (Combellick and Osborne, 1977) and

- 25 more than three times as much as in coastal dunes in Baja California to the south (Kasper-Zubillaga et al., 2007). This mineralogically immature sand suggests rapid erosion of a feldspar-rich source rock, potentially derived from the Santa Maria River drainage. More mineralogically mature terrestrial dune fields, including most old and continental-based (as opposed to coastal) terrestrial desert sand seas and some older coastal dunes, are quartz-dominated and feldspar-depleted (Brookfield and Ahlbrandt, 1983; Pye and Tsoar, 2009; Swet et al., 2019). The removal of feldspars by ballistic-impact-derived breakdown
- 30 into dust during aeolian recycling over long periods of time contributes to the mineralogical maturation (into quartz-dominated deposits) of aeolian dune sands (Muhs et al., 1997; Muhs, 2004).

Furthermore, weathering of feldspars to clay minerals might produce some of the clay-mineral coatings on sand grains (O'Hara-Dhand et al., 2010) at the study site. Feldspars in the Oceano sand, especially plagioclase, are vulnerable to chemical weathering under the humid and salt-bearing coastal conditions (James et al., 1981; Pye and Tsoar, 2009) and mechanical breakdown (aeolian abrasion) into dust from saltator impacts in wind events (Muhs et al., 1997). Additionally, Compton (1991)

5 described how the Santa Maria basin source sediments, both onshore and offshore, contained very fine clays derived from the alteration of volcanic glass mediated by and enhanced by weathering of feldspars. As a result, the combination of abundant feldspars in the Oceano sand and clay minerals from an additional source could help explain the observed clay-mineral coatings on Oceano sand and the fine-grained characteristics of Oceano dust emissions.

Our results also imply that the dust-emission processes at Oceano are not main contributors to dust emissions from North Africa. In situ measurements of the PSDs of atmospheric dust over North Africa are significantly coarser than the PSD of dust emitted from our Oceano site (Fig. 3B) and the PSD of dust generated in laboratory experiments on the removal of clay-mineral coatings (Fig. S10B; Bullard et al., 2004; 2007). As such, although the processes responsible for dust emission from the Oceano sand sheet - removal of clay-mineral coatings and aeolian abrasion of feldspars - could be important for specific North African dust sources, they are likely not primary dust emission processes on a regional scale in North Africa.

## 15 **4.3** Implications of dust emissions from Oceano Dunes State Vehicular Recreation Area for downwind human health, park management, hydrological cycle, and climate

Dust emitted from our Oceano site is significantly finer than dust emitted from non-sandy soils (Fig. 3A), which can amplify its impacts on downwind human health, the hydrological cycle, and climate. Fine dust has a longer lifetime, and therefore is usually transported over longer distances than coarse dust (Mahowald et al., 2014). Furthermore, fine dust, especially within

20 the PM<sub>2.5,a</sub> size range, is associated with cardiopulmonary diseases, lung cancer, and ischemic heart disease (Burnett et al., 2014). On a per mass basis, dust emitted from Oceano, and from other source regions with similar emission processes, can thus be expected to have greater risks to downwind human health than dust emitted from non-sandy soils.

Our measurements of dust emission from the Oceano Dunes State Vehicular Recreation Area (SVRA) could help inform decisions on the management of this California state park. Our measurements were performed in the part of the park where

- 25 public off-highway vehicle (OHV) use is prohibited and we found that substantial dust emissions occur in the absence of OHV use (Fig. S1). Because OHV use can enhance dust emissions (Goossens and Buck, 2011; Goossens et al., 2012), reconfiguring motorized access to the park could modify dust emissions and associated downwind dust impacts on human health, vegetation, and water quality (Ouren et al., 2007). Measurements by Gillies et al. (2017) (see their Fig. 10) indicated a positive relationship between airborne PM<sub>10</sub> and wind speed in a section of Oceano Dunes SVRA with vehicular activity, however, their
- 30 measurements of riding area dust concentrations are not directly comparable to our calculated non-riding area dust emission fluxes. Disentangling the relative contributions of natural and OHV-influenced dust emissions will be essential as Oceano

Dunes SVRA responds to a dust emission abatement order recently approved by the local Air Pollution Control District Hearing Board (Vaughan, 2018).

Dust emitted from Oceano sands possibly contains a large proportion of feldspars, and thus might modify downwind cloud microphysics and hydrology. Oceano sand has ~46% feldspars by mass (Table 2), which is on the high end for dust-source

- 5 regions (Murray et al., 2012; Atkinson et al., 2013). Consequently, the content of feldspars in dust emitted from Oceano is likely on the high end as well, especially considering that aeolian abrasion of feldspars (Muhs et al. 1997) could be one of the emission processes. Feldspars, especially K-rich feldspars, are considered the most important ice nuclei for mixed-phase clouds, based on both laboratory (Atkinson et al., 2013) and in situ measurements (Price et al., 2018). Recent observations suggest that cloud glaciation mediated by dust aerosols contributes to more than half of ice-phase precipitation in the Sierra
- 10 Nevada mountain range (Creamean et al., 2013), which is a major source of water for California residents (Dettinger et al., 2004). Because Oceano sands are rich in feldspars, we speculate that dust emitted from Oceano could potentially contribute to glaciating downwind clouds, thereby affecting precipitation in the Sierra Nevada region. Future measurements to determine the broader occurrence of dust production by feldspar abrasion for active sands with high feldspar content are thus clearly needed.
- 15 Our findings that dust emitted from Oceano sands is relatively fine, and that the dust flux is on the low end of that emitted from non-sandy soils, indicate the need for substantial further study to understand the role of dust emissions from active sands in past, current, and future climates. Partially because fine dust has a longer lifetime and larger surface area per unit mass than coarse dust (Mahowald et al., 2014), it produces a substantial cooling radiative effect (Kok et al., 2017). Therefore, it is important to include size-resolved dust emission in climate studies. However, the parameterization of dust emissions from
- 20 active sands in climate models is difficult because of a scarcity of measurements of dust emissions from active sands. Although our measurements help to fill this gap, it remains unclear how representative our measured fine dust emissions from coastal Oceano sands are for typical active sands. Therefore, there is a clear need for future investigations of dust emissions from active sands for a range of geomorphological and sand properties, including grain size distribution, mineralogy (such as extents of clay-mineral coatings and feldspars), chemical and physical weathering rates, dune type, and palaeoenvironmental history.
- A further complication in representing the role of dust emissions from active sands in climate models is that sand dunes can transition between stable and active states in past, current, and future climates. Many stable dunes have accumulated extensive clay-mineral coatings (Bowler, 1973; Gardner and Pye, 1981; Muhs et al., 1997), and, if activated, these dunes could possibly produce substantial fine dust emissions through sandblasting-induced removal of clay-mineral coatings (Fig. S10; Bullard et al., 2007; Swet et al., 2019). The transition from stable to active sand dunes can be triggered by increases in wind speed or
- 30 decreases in vegetation coverage, which in turn can be caused by decreased precipitation or by human activities, such as grazing and land-use change (Ashkenazy et al., 2012). Areas with coexisting active and stable sand dunes, such as coastal zones, are especially vulnerable to the reactivation of stable sand dunes (Yizhaq et al., 2009), and could thus become potent

dust emission sources, especially if the dunes experienced some weathering in the humid, salt-rich coastal environment (Muhs et al., 1997). Because active sands are generally not represented as dust emission sources in climate models, reactivation of sand dunes might thus enhance future dust effects on climate beyond what current models simulate (Kok et al., 2018).

#### 4.4 Limitations of the methodology

- 5 Our methodology has important limitations. First, our XRPD analysis could not distinguish the concentrations of clay-mineral coatings from loose individual fine particles contained in the pore spaces between sand grains. This uncertainty limits our interpretation of the relative contributions of removal of clay-mineral coatings and saltation-driven release of loose fines to our measured dust fluxes. Second, we did not collect aeolian dust samples simultaneously with the field measurements. Instead, we collected soil samples on October 14<sup>th</sup>, 2016, more than a year after the field measurements in the summer of 2015, such
- 10 that it is possible that these samples are not representative of the sand surfaces at the time of our field campaign. However, sand-surface properties over a large area are unlikely to change substantially within a timescale of one year (Hillel, 1998). Third, we approximated dust as spherical particles when correcting the OPC bin sizes, although dust is highly aspherical (Okada et al., 2001; Kandler et al., 2007). Because OPC measurements use side-scattered light to determine dust size, which remains poorly understood (Nousiainen and Kandler, 2015), we followed past work (Sow et al., 2009; Rosenberg et al., 2014;
- 15 Ryder et al., 2018) in approximating dust as spheres. Fourth, we did not quantify the effect on the measured PSD due to the particle size- and wind speed-dependent sampling efficiency (Von der Weiden et al., 2009), which might be important especially for the largest particle bin (bin 6), as suggested by the decrease in sandblasting efficiency with shear velocity for this bin, which contrasts with the increase with shear velocity for the smaller particle bins. Therefore, it is possible that the PSD of emitted dust (Fig. 4) remains invariant with shear velocity.

#### 20 5 Conclusion

We presented the first (to our knowledge) in situ field measurements of dust emissions under natural saltation from an active sand sheet at the coastal Oceano Dunes in California (Figs. 1 and S1). We found that although the  $PM_{10,g}$  dust emitted from Oceano is on the low end of the range in emissions produced by non-sandy soils for similar wind speed (Fig. 2A), it is also substantially finer (Fig. 3A) such that the  $PM_{2.5,a}$  flux is comparable in magnitude to dust emitted from non-sandy soils (Fig.

25 2B). As fine dust has a longer atmospheric lifetime, is associated with adverse impacts on human health, and produces a cooling radiative effect, dust emissions from Oceano could impact downwind climate and human health.

Our results provide insights into the physical processes that drive dust emissions from the sand sheet at Oceano Dunes. We found that Oceano sand has substantial clay-mineral coatings and feldspars (Table 2), suggesting that dust is emitted through a combination of removal of clay-mineral coatings, release of resident fines, and ballistic breakdown of feldspars by saltation

impacts. As feldspars are the most efficient ice nuclei for mixed-phase clouds, we speculate that dust emitted from Oceano could potentially contribute to glaciating downwind clouds, thereby affecting precipitation in the Sierra Nevada region.

Our measurements provide limited insights into the contribution of active sands to the global dust cycle. We found that the PSD of dust emitted from Oceano is much finer than observed in situ over North Africa (Fig. 3B). Therefore, although the

- 5 processes responsible for dust emission at our Oceano site removal of clay-mineral coatings and aeolian abrasion of feldspars - could be important for specific North African dust sources, they are likely not primary dust-emission processes on a regional scale in North Africa. However, because it is unclear how representative the dust-emission processes at our Oceano site are for emission processes occurring at typical active sands, the contributions of active sands to dust emission from North Africa and other globally-important source regions remain uncertain. Further work on dust emissions from active sands with various
- 10 properties (sand size distribution, mineralogy, chemical and physical weathering rates, dune type, and palaeoenvironmental history) is thus needed to evaluate the contribution of active sands to the global dust cycle.

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Table 1: The dust geometric and aerodynamic diameters of the seven bin boundaries. Determination of the geometric diameter ranges uses Lorenz-Mie theory such that they produce the same range of scattered light intensity as the seven manufacturer-provided polystyrene latex sphere (PSL) diameter sizes. The ranges in geometric diameters derive from the uncertainty in the dust refractive indices. We
 determined the aerodynamic diameter ranges by applying Eq. (6) to the geometric diameter ranges.

Polystyrene latex sphere (PSL) diameter (µm)	Dust geometric diameter (µm)	Dust aerodynamic diameter (µm)
0.49	0.51 - 0.54	0.68 - 0.72
0.7	0.73 – 0.77	0.97 – 1.03
1	1.01 – 1.03	1.35 – 1.37
2	2.18 - 2.32	2.91 - 3.09
2.5	2.67 – 2.90	3.56 - 3.87
5	6.11 – 6.31	8.15 - 8.41
7	8.89 - 9.40	11.85 – 12.53

15 Table 2: Averaged mineralogy of the Oceano Dunes sand samples in unit of percentage by mass, determined from X-ray powder diffraction (XRPD) analysis. Note that the values in the table are averages of mineralogy of four 1-gram samples with two collected from the tower location and two from 100 meters upwind. Each analysed sample contains hundreds of grains.

	Quartz	Potassium feldspar	Sodium-rich	Clay minerals
	-	(Microcline and	plagioclase	(Illite)
		Orthoclase)	feldspar (Albite)	
Oceano Dunes	51	23	23	3



**Figure 1: The experimental setup at Oceano Dunes.** Instrumentation includes six sonic anemometers (S1-S6), nine Wenglor particle counters (W1-W9), eight Big Spring Number Eight sand samplers (A1-A4, B1-B4), four optical particle counters (D1-D4), and a weather station with temperature and humidity sensors (WS) (after Martin and Kok, 2017).



Figure 2: The vertical dust mass flux as a function of shear velocity within (A) the  $PM_{10,g}$  geometric diameter range and (B) the  $PM_{2.5,a}$  aerodynamic diameter range. Each of the two plots includes measurements at Oceano (black open circles), and previously published field studies of natural dust emissions from non-sandy soils (open triangles). These latter measurements were compiled in Kok et al. (2014a), which corrected these measurements to the  $PM_{10,g}$  geometric diameter range following the procedure described in that work. Furthermore, we corrected these measurements to the  $PM_{2.5,a}$  aerodynamic diameter range assuming that their PSDs follow the prediction of brittle fragmentation theory generated by aggregate fragmentation (Kok, 2011). Error bars on the Oceano measurements were obtained through error propagation (details in Supplement).

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10 Figure 3: Normalized volume particle size distribution (PSD) of dust at emission as a function of dust aerosol geometric diameter averaged over all measurements at Oceano (black open circles). Plotted for comparison are measurements compiled by Kok et al. (2017) of (A) the emitted dust PSDs from non-sandy soils in the U.S. (Gillette et al., 1972; 1974; Gillette, 1974), China (Fratini et al., 2007), North Africa (Sow et al., 2009; Rosenberg et al., 2014), and Australia (Shao et al., 2011), and (B) in situ aircraft measurements of dust-dominated PSDs close to the Saharan source regions. Note that these measurements are normalized using the procedure described in Kok (2011) and

15 Kok et al. (2017), which differs somewhat from the procedure used for the Oceano data (see Methods). Also plotted for comparison is the brittle fragmentation theory (blue dash-dotted lines) on the PSD of emitted dust generated by aggregate fragmentation (Kok, 2011). Error bars on the Oceano measurements were obtained through error propagation (details in Supplement).



Figure 4: Shear velocity dependence of the normalized volume particle size distribution of dust at emission at Oceano. We divided the measurements evenly into three shear velocity bins, with averaged values of 0.33, 0.37 and 0.41 m/s, respectively. Error bars on the measurements were obtained through error propagation (details in Supplement).





Figure 5: The bulk sandblasting efficiency  $\alpha$  (m<sup>-1</sup>) at Oceano as a function of shear velocity. The linear-least squares fit (black line) indicates that  $\alpha$  increases as a power law in shear velocity with an exponent of 3.50±0.89. Error bars were obtained by propagating errors from the sensor intercalibration process, the regression used in the gradient method, and the subtraction of the sea-salt deposition flux (see Supplement). The two black dotted lines denote the 95% confidence range on the fit. The geometric and the aerodynamic diameter ranges

of the vertical dust mass flux  $F_d$  are noted at the top, and Q is the total horizontal sand flux integrated over all sand grain sizes (Martin et al.,

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2018).



Figure 6: The sandblasting efficiency  $\alpha_i$  (m<sup>-1</sup>) of the six size bins at Oceano as a function of shear velocity. The linear-least squares fit (black line) indicates that  $\alpha_i$  varies as a power law in shear velocity with an exponent of 4.71±0.97, 5.00±1.37, 5.22±1.63, 6.12±2.11, 3.26±1.90, and -2.89±1.94 for the six respective size bins. Error bars were obtained by propagating errors from the sensor intercalibration process, the regression used in the gradient method, and the subtraction of the sea-salt deposition flux (see Supplement). The two black dotted lines denote the 95% confidence range of the fit. The geometric and the aerodynamic diameter ranges of the vertical dust mass flux  $F_{d,i}$  are noted at the top of each of the six plots, and Q is the total horizontal sand flux integrated over all sand grain sizes (Martin et al.,

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2018).



	Oceano
Statistical parameters (µm)	
Mean	538.28
Mode	461.48
D10	270.61
D50	490.95
D90	879.67
Weight fraction (%)	
Clay-sized (< 2 μm)	0.16
Clay- and Silt-sized (< 63 µm)	0.95
Fine sand (63-250 µm)	7.93
Meidum sand (250-500 µm)	54.35
Coarse sand (> 500 μm)	44.69
PM <sub>1</sub> (< 1 μm)	0.09
PM <sub>2.5</sub> (< 2.5 μm)	0.19
PM <sub>10</sub> (< 10 μm)	0.41

**Figure 7: Normalized volume particle size distribution (PSD) of sand samples collected from the Oceano field site.** Statistical parameters of the PSD are reported in the table to the right. D10, D50 and D90 refer to cut-off diameters of the 10<sup>th</sup>-, 50<sup>th</sup>-, and 90<sup>th</sup>-percentile of sample grains by mass, respectively. Note that the values are averages of four samples with two collected from the tower location and two from 100 meters upwind.

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Figure 8: (A) Scanning Electron Microscopy (SEM) image of an Oceano sand sample. (B) Energy Dispersive X-ray Spectroscopy (EDS) chemical composition (percentage by mass) of a quartz grain in image A (the location of the analysis is marked by the black letter "B"). (C) EDS chemical composition of a K-rich feldspar in image A marked by "C". (D) Close-up image of a quartz sand grain with mineral coatings. (E) EDS chemical composition of the mineral coatings in image D marked by "E".