

Insights into the single particle composition, size, mixing state and aspect ratio of freshly emitted mineral dust from field measurements in the Moroccan Sahara using electron microscopy

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General Comments

10 The manuscript describes the sampling and analysis of mineral dust samples collected in the Moroccan Sahara in September 2019. For inter-comparison the dust particles were collected by three sampling methods, including flat-plate sampler (FPS), free-wing impactor (FWI), and a multi-stage deposition impactor (MOUDI). Real time particle size distributions were also measured by optical particle counter (OPC). The samples were analyzed by automated scanning electron microscopy (SEM), generating analyses on particle morphology and

15 chemical composition, of more than 300,000 individual dust particles. Normalized elemental results were assembled into eight particle classes by a set of rules, each class approximating a likely dust mineral. These were labelled as hematite-like, quartz-like, feldspar-like, calcite-like, gypsum-like, halite-like ammonium sulphate-like, kaolinite-like, etc. The results were classified by particle diameter into eight size bins from, < 1 to 128 μm , each size bin containing the particle counts in that size bin. Inter-comparative plots of number

20 abundances of the mineral-like and other chemical parameters abundances are provided. Discussion of the iron comparative number distribution plots for the FPS, FWI, two stages of the MOUDI, as well as for the OPC are given, with discussion of the mixing state thereof. Aspect ratios are compared with those from other studies.

The title clearly reflects the content of the paper and the abstract provides a concise summary.

25 It adequately addresses the *in situ* sampling and SEM analysis of mineral dusts, of importance to a better understanding of airborne minerals and their potential impact on Global climate.

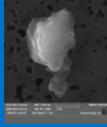
Although automated SEM analysis has been known for more than two decades (Engelbrecht et al., 2009), the application thereof to dust analysis remains novel, especially regarding the interpretation of data. Research

presented in this paper further explores the automated SEM procedures and technology. A Global library of
30 airborne dust mineralogical and morphological data will facilitate the interpretation of satellite signals. To
readily compare with other similar studies, the chemical results should be calculated to reflect mass fractions or
mass percentages, not number abundances.

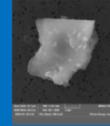
Specific Comments

35 1. Particle **number** abundances (fractions) per “volume equivalent diameter” are presented in figures and
text in the manuscript. In a few instances (lines 16, 447, 532, 574) the authors do refer to volume
fractions. As shown in the below figures (Engelbrecht et al., 2017), there is a substantial bias towards
the number of smaller particles. To better compare with other bulk analytical techniques, individual
particle **volume** or **mass** (Ott and Peters, 2008a; Prakash et al., 2016; Engelbrecht et al., 2009;
40 Engelbrecht et al., 2017; Marsden et al., 2018; Marsden et al., 2019; Ott and Peters, 2008b) can be
assessed from SEM based particle area measurements (Engelbrecht et al., 2009; Prakash et al., 2016;
Ott and Peters, 2008b). Similarly, for intercomparison of chemical parameters between particle size
bins, it is preferable to express abundances as equivalent **volumes**, and if particle densities are known,
particle mass.

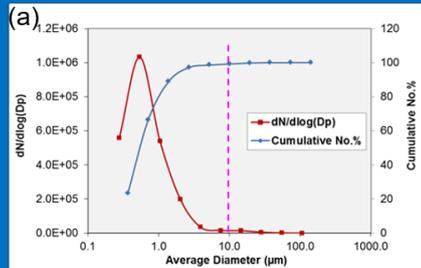
45 It is recommended that the number concentrations be converted to at least volume concentrations
throughout this manuscript, and that the chemical parameters be assessed in terms of volume fractions
or volume percentages. The figures illustrating the chemical parameters will need to be re-compiled to
reflect volume (or mass) concentrations and not number abundances. In this way the results can be
readily compared to other SEM based studies, as well as to chemical results generated by other
50 analytical techniques such as SPMS, ICP-MS, XRF, etc.



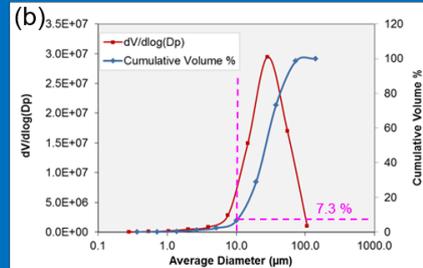
Dust particle size distribution by SEM



Number concentration



Volume concentration



Particle size distribution of sample collected by inverted "Frisbee" sampler on KAUST campus in December 2015, by (a) number, and by (b) volume, of about 2,000 particles measured by scanning electron microscopy (SEM)

2. Results from this study should be discussed in conjunction with individual particle dust studies from other desert regions, including North Africa and the Middle East (Engelbrecht et al., 2009; Engelbrecht et al., 2017; Prakash et al., 2016).

55

Some Technical Corrections to consider

Line 43. properties and mineral composition

Line 47. deposit **faster** due to gravity,

60 Line 53. ..dust is more **optically** absorbing when...

Line 57. ...not only on the source **composition** but also on the feldspar...

Line 80. ...are **the interdependencies among particle** size, composition,...

Line 82. ...mixing state upon both **particle** composition and size...

Line 83. ...mineralogical composition of **windblown dust** are relatively new ...

65 Line 89. Unclear, rephrase sentence. "One reason for such measurement scarcity is that source areas, particularly the host prolific ones, are typically located in remote areas and subject to harsh conditions."

Line 91. Dust storms result in high particulate concentrations and filter overload, providing a challenge to analyze by automated SEM.

Line 96. Three different instruments were used to sample mineral dusts, for the measurement of chemical and physical properties of individual dust particles by SEM coupled with an energy-dispersive X-ray analyzer (EDX).

70

Line 99. ...individual dust minerals to ultimately help understand...

Line 101. Such information is required to better constrain climate models that consider mineralogical variations in their representation of the dust cycle (Perlwitz et al., 2015a; Scanza et al., 2015; Li et al., 2021). This is timely, given the prospect of global soil mineralogy retrievals using high-quality satellite hyperspectral measurements

75 (Green et al., 2020).

Line 117. Under favorable weather conditions, dust is frequently emitted from this source area. (González-Flórez et al., in prep.).

Line 129. ...FWI were collected twice daily with a typical....

Line 138. ...particle size cutoff is determined by the impaction ...

80 Line 145. The main controls for particle ...

Line 153. ...and fourth stages were selected for detailed analysis.

Line 154. ...when particles impact on the collection substrate....

Line 156.depending on the mineralogical properties of particles....

Line 162. Previous studies (Kandler et al., 2008.....

85 Line 162. Add references (Engelbrecht et al., 2009; Engelbrecht et al., 2017; Prakash et al., 2016)

Line 166.without any pretreatment. Particles were automatically detected and analyzed

Line 167. ...(BSE) imagery was used for particle detection, as dust particles

Line 170. ... number of input counts by the EDX detector.

Line 174. Accounting for matrix dependent efficiencies of the....

90 Line 176. The SEM-EDX results are normalized to 100 %, including those of the elements C, N, O, Na, Mg, Al, Si, P, S, Cl, K, Ca, V, Cr, Mn, Fe, Zn, and Pb.

Comment: What is the minimum particle size measured by automated SEM-EDX?

Line 228. SEM-EDX measures the elemental composition

Line 229. ...can therefore be readily identified (e.g. gypsum, quartz, calcite).

- 95 Line 238. X is defined as the ratio of the **mass** concentration of the element considered to the sum of the **mass** concentrations of all the measured elements.
- Line 243. ...method **was not applied** to quantify...
- Line 246. Their most prevalent **chemical** components, ...
- Line 247. **The mineral labels were assigned from the most prevalent measured elemental concentrations. There**
- 100 **was no actual phase** identification
- Line 252. Aerosol PSDs of suspended mineral dusts were **also obtained by** Optical Particle Counter (OPC).....
- Line 264.only error considered is the Poisson counting error. **Explain or delete sentence**
- Line 268 Based on **set classification rules, particles**....
- Line 275 ...particle types were **found mainly** in particles with...
- 105 Line 284.Fe content of quartz-like particles is generally **low and variable, suggesting that Fe oxides are not an integral part of the quartz-like particles.**
- Line 289. Ca-feldspar-like particles are quite rare in the samples. **Explain why, plagioclases are seldom pure albite, often oligoclase or andesine, depending on their provenance. This anomaly perhaps to do with the SEM analytical procedure?**
- 110 Line 301. Figure 2 y-axis. **Are relative abundances by number or volume (or mass)?**
- Line 326. Ammonium sulfate is the prevalent **sulfate species in atmospheric aerosols and generally of anthropogenic origin.**
- Line 373. On average across all particle sizes, **by number**, 26% of particles are illite-like,
- Line 416. along the **air stream onto the lower** stages.
- 115 Line 422. While the dependence of particle size distribution on **sample mineralogy is quite** strong, the temporal ...
- Line 431. **There are fertilizers that do not contain K, such as di-ammonium phosphate (DAP), mono-ammonium phosphate (MAP). Hydroxy apatite is not commonly found in the Morocco phosphate deposits, the mined francolite generally contains F.**
- 120 Line 439. Figure 8. **Phase compositions of size bins for samples collected under specific meteorological conditions at specific times and on certain days.**
- Line 439. The total number of analyzed particles **is** given for.....
- Line 441. **feldspar not a typical Fe bearing mineral, only K-feldspar in very small amounts.**

Line 474, Figure 9. **Relative number abundances?**

125

Engelbrecht, J. P., McDonald, E. V., Gillies, J. A., Jayanty, R. K. M., Casuccio, G., and Gertler, A. W.: Characterizing mineral dusts and other aerosols from the Middle East – Part 1: Ambient sampling, *Inhalation Toxicology*, 21, 4, 297-326, doi: 10.1080/08958370802464273, 2009.

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Ott, D. K., and Peters, T. M.: A Shelter to Protect a Passive Sampler for Coarse Particulate Matter, *PM_{10-2.5}*, *Aerosol Science and Technology*, 42, 4, 299-309, 10.1080/02786820802054236, 2008.

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