

**1) Editor comment:** I can't find the following change in the revised manuscript.

Reviewer #2: Pg 3/line 14: It is not really accurate to refer to the "IN fraction". A fraction of particles can be IN active at a /specific/ temperature. Most mineral phases are IN active at some mixed-phase cloud temperature, even weak ice nucleants such as quartz. Perhaps rephrase to refer to the ice nucleation properties of mineral dust particles (which requires knowledge of the specific mineral phases present).

This sentence has been re-written to: "However, relating IN properties to mineral phase in natural dust particles is much more difficult due to complex mineralogy and mixing state that is difficult to resolve."

**2) Editor comment:** In your response below, it seems that you intended to add a discussion of the efficiencies. Was this actually done?

Reviewer #2: 5/5: Just referring the reader to another paper for relevant information is not satisfactory. Please provide a clear summary here regarding the relevant performance characteristics of this LAAPTOF configuration that influences what fraction of particles are actually detected, put in the context of the aerosol populations analyzed here and their associated properties that govern particle sample and detection.

We have added a paragraph that summarises performance characteristics previously reported with this instrument with mineral dust:

"Laboratory evaluation of the fiber-coupled laser system indicate that the detection efficiency peaks at 0.25 with spherical particles {Marsden2016a}, but the overall efficiency of the instrument also depends on ablation efficiency with respect to particle composition. In a study of nominally pure mineral samples, {Marsden2017} reported the number of optically detected particles that produced a mass spectra (i.e. hit rate or ablation efficiency) of 0.29 and 0.14 for illite and kaolinite respectively, but was also dependent on the amount of impurities such as Titanium. Furthermore, from the authors own experience, it likely that pure quartz may have an ablation efficiency close to zero, so that the potential range of overall efficiency ranges from 0.0725 for spherical particles of

illite, to almost zero for pure quartz particles. The exact efficiency of the instrument is not known in most situations because the size, shape and composition of the particles would have to be known a priori.”

A discussion of the impact of these efficiencies on the current measurement are discussed in....

**3) Editor comment:** The reviewer commented on your sentence ‘Similarly, organic markers C<sub>2</sub>;C<sub>2</sub>H and C<sub>2</sub>H<sub>2</sub>(m=z24;25;26) appear on particles after mixing suspended feldspar with ozone and  $\alpha$ -pinene (Fig. 4(c)). (now p. 8, l. 13) as below. Your response does not match the reviewer’s comment as your wording still implies that C<sub>2</sub> can be used as an organic marker.

Reviewer #2, 7/10: Also, while organics can “char” to EC ions, C<sub>2</sub><sup>+</sup> could also come from black carbon that was mixed with the particle. I would be wary of using C<sub>2</sub><sup>+</sup> to identify organic carbon, it is a generic carbonaceous (OC + BC) marker.

We do not actually use the C<sub>2</sub> marker to identify organics, we are just pointing out which ion combinations appear in the spectra after the addition of organic material.

**4) Editor comment:** The following comment has not been addressed at all. (now: P. 13, l. 25)

Reviewer #2: 12/23: Uptake of HCl by dust observed using SP-MS was a focus of (Sullivan et al., 2007b), which you cite yet oddly do not discuss when very relevant here.

**5) Editor comment, follow-up on Reviewer #3:** Abstract, line 19-20: please clarify the sentence now: p. 1, l. 22-3: This sentence still sounds strange since the subject in the two fragments changes and it is not clear what a ‘continuous difference’ is.

Maybe better something like:

In most cases, the differences in the mineralogical composition between particles within a soil sample were small. Thus, particles were not composed of discrete mineral phases.

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## Minor editor comments

p. 3, l. 30: consist

p. 3, l. 35: of these cations...

p. 3, l. 35: charge-balanced

p. 4, l. 3: to be abundant

p. 4, l. 7: is chemically similar...

p. 4, l. 24: either 'shows' or '...these techniques show evidence...'

p. 4, l. 28: describes → describe

p. 4, l. 29: semicolon is redundant

p. 5, l. 12: 'concentration is...' or 'concentrations are...'

p. 5, l. 12: 'achieved' does not seem to be the right word here. Replace by 'observed' or 'detected'

p. 5, l. 13, and l. 16: These techniques...

p. 6, l. 29: either 'produced a mass spectrum' or 'produced mass spectra'

p. 6, l. 30: authors' own ...

p. 6, l. 31: verb missing (it is likely...(?))

p. 6, l. 33: 'particles' is redundant

p. 8, l. 27: Is it really SO<sub>4</sub><sup>-</sup> or rather SO<sub>4</sub><sup>2-</sup>?

p. 8, l. 29: it is therefore...

p. 9, l. 15: per sample

p. 9, l. 19: skew and suppress

p. 9, l. 20: remove the last 'ratios'

p. 11, l. 17: ...are dynamic

p. 12, l. 16; p. 13, l. 29: Is it really Ca<sup>+</sup> or Ca<sup>2+</sup>?

p. 12, l. 12: due to...

p. 14, l. 18: are much reduced...

p. 14, l. 23: consideration

p. 14, l. 29: which section are you referring to?

p. 17, l. 16: measurements show

p. 17, l. 27: either '...numbers suggest' or 'number suggests...'

Figure 9, caption: should this read  $0.58 < \tau < 0.8$ >

Figure 9: Add a) , b) ... to the individual figures

Figure 12, caption: “A total of 12698 (a) concentration of silicate and calcium rich particles (total measurement of 12698 silicate and 6837 calcium rich particles).” - Please reword and clarify.

Figure 13, caption: - (1301 dust particles analysed)

- back trajectory analysis suggests

References : Ahern et al : Cite AMT paper instead of AMTD