Responses to Jonathan Merrison's comments (RC1):

Thanks are extended to the editor, Paola Formenti, and to the reviewers, Jonathan Merrison and an anonymous reviewer, for their careful work and thoughtful suggestions that greatly improved the manuscript.

The following text contains the reviewer's comments (black), our replies (blue) and the changes made to the manuscript (red).

Comment 01: This paper presents the results and analysis of a field campaign studying the electric field generated by a series of dust storms. This work is of high interest to those studying Aeolian processes, atmospheric electricity, aspects of planetology and especially the electrification of aerosols and granular materials. This is one of only a few such studies and it appears to be thorough and in depth applying a range of experimental sensor techniques.

Response:

We thank the reviewer for this positive assessment of our manuscript.

Comment 02: Generally in this paper the distinction between sand and dust is unclear, an example is line 155 in which the size distribution of saltating particles (by definition these must be sand) are used to make arguments about dust events. By definition sand grains cannot be suspended and are transported at low altitude (typically <1m), cohesion effects for sand are typically negligable. Dust cannot saltate and it is unclear whether they can/do in fact collide while in suspension, it may be that if in contact dust grains will cohere (aggregate). It is my impression that this study focusses on atmospheric dust in which case this should be made clear.

Response:

As you pointed out that our study is mainly concerned with airborne dust particles, but in the original manuscript, the size distributions of saltating particles rather than airborne dust particles are used to describe the observed dust storms. In the revised manuscript, we have added the measured size distributions of airborne dust particles collected at the S9 site (5 m above the ground), and we can see that dust events occurring in the QLOA site have a very similar dust particle size distribution. The related sentence in the revised manuscript has been modified as:

Measurements of the size distribution of airborne dust particles (Fig. S2) and saltating particles (Fig. S3) implies that the dust events occurring in the QLOA site have a very similar particle size distribution.

In addition, the size distributions of airborne dust particles have been provided in Fig. S2 in the Supplement, as follows:



Fig. S2. Size distributions of the airborne dust particles collected at the S9 site (5 m above the ground). (a) A dust collector was mounted on a horizontally orientated steel bar. (b) Number distribution of the collected airborne dust particles during No. 01 and No. 02-10 dust storms. (c) The corresponding volume distribution of the collected airborne dust particles. Particle size analysis was performed using the Microtrac S3500 tri-laser particle size analyzer. Since the collected airborne dust particles of single dust storms are very few (i.e. No. 02-10 events), it is difficult to measure the size distribution of single dust storms by the collected dust sample. Consequently, the collected dust particles from No. 02-10 dust storms were combined to obtain a mean size distribution, as shown in Figs. S2a and S2b.

Comments 03: It is of some interest whether the field measurements made here are consistent with dust being typically electrified negatively, the authors might want to comment upon this.

Response:

In our field campaign, the observed space charge density at 5 m height is positive, which is consistent with the previous studies such as Kamra (1972) and Williams et al. (2009). Actually, the charge structure of dust storms is generally multipolar. In the revised manuscript, we have discussed this topic in detail as follows:

Previous measurements have demonstrated that the charge structure of dust clouds in dust storms could appear as unipolar, bipolar, and even multipolar. For example, Williams et al. (2009) measured the vertical E-field in dust storms and found both upward- and downward-pointing vertical E-field. They inferred that the dust cloud is unipolar if the near-ground particle charge transfer is dominating, while the dust cloud is bipolar if upper-air (volume) charge transfer is dominating. Direct dust storm charge measurements by Kamra (1972) have also observed both positive and negative space charge at 1.25 m height above the ground. Additionally, our recent dust storm E-field measurements up to a height of 30 m have shown that dust cloud could be multipolar (Zhang et al., 2017). In this study, the derived space charge density at 5 m height is positive, which is certainly reasonable, although many studies have observed a negative space charge. In fact, the charge structure of dust storms is closely associated with the transport of dust particles. There is no doubt that the large-scale and very-large-scale motions of flow exist in the high Reynolds number atmospheric surface layer (Hutchins et al., 2012), affecting the transport of dust particles because of dust following wind flow exactly (Jacob and Anderson, 2016). We can expect that a bipolar charge structure in each large-scale motions is produced by the bi-disperse suspensions of oppositely charged particles (Renzo and Urzay, 2018). Consequently, the multipolar charge structure of dust storms is formed by a series of bipolar charge of large-scale motions.

References:

- Hutchins, N., Chauhan, K., Marusic, I., Monty, J., and Klewicki, J.: Towards reconciling the large-scale structure of turbulent boundary layers in the atmosphere and laboratory, Boundary-Layer Meteorol., 145, 273-306, <u>https://doi.org/10.1007/s10546-012-9735-4</u>, 2012.
- Jacob, C., and Anderson, W.: Conditionally averaged large-scale motions in the neutral atmospheric boundary layer: Insights for aeolian processes, Boundary-Layer Meteorol., 162, 21-41, <u>https://doi.org/10.1007/s10546-016-0183-4</u>, 2016.
- Renzo, M. D., and Urzay, J.: Aerodynamic generation of electric fields in turbulence laden with charged inertial particles, Nat. Commun., 9, 1676, <u>https://doi.org/10.1038/s41467-018-03958-7</u>, 2018.

Comments 04: The consecutive acquisition of charge (Line 73-80) through collisions and a so called equilibrium charge has been observed only in some laboratory studies (mostly using sand sized particles), it is not demonstrated in all experiments and is not generally accepted that electrification of dust in fact involves multiple collisions. Recent laboratory work implies that it is not (e.g. Alois et al. 2017, 2018). Similarly it seems that the work presented here is not in fact in agreement with a model based upon multiple collisions (e.g. line 259, 406). (Alois, S., Merrison, J., Iversen, J.J., Sesterhenn, J., (2017) Contact electrification in aerosolized monodispersed silica microspheres quantified using laser based velocimetry, Journal of Aerosol Science 106 1–10., Alois, S., Merrison, J., Iversen, J.J., Sesterhenn, J., (2018), Quantifying the contact electrification of aerosolized insulating particles, Powder Technology 332, 106–113)

Response:

We are sorry for our negligence of recent important studies associated with contact electrification of micron-sized particles (e.g. Alois et al. 2017, 2018). Alois et al. (2017) simultaneously measured the size and electrical charge of individual micron particles using a novel technique based on laser velocimetry and found that the surface charge density of the charged particles closely distributed around a constant value of 0.02 mC m⁻², which was probably caused by the electron field emission at particle contact site. According to the reviewer's suggestion, the changes made in the revised manuscript are threefold:

(i) line 73-83 has been modified as follows:

Furthermore, a large number of theoretical work and laboratory experiments have shown that the net electrical charge on millimeter-sized particles increases with increasing number of collisions/contacts and correlates with the particles' kinetic energy (Apodaca et al., 2010; Harper and Dufek, 2016; Matsuyama and Yamamoto, 1995; Zhang et al., 2013) until particles acquired a certain amount of charge (termed as equilibrium charge). The magnitude of the eventual equilibrium charge on particles is found to be independent of the particles' collisional dynamics and therefore will reduce the difficulties in the quantification of particle electrification. However, whether such an electrification equilibrium exists under natural circumstances, especially for micron-sized dust particles in dust storms, is unclear and needs to be verified.

(ii) The references of Alois et al. (2017, 2018) have been cited in the revised manuscript. That is, in section 4.2 we have added the following text:

More recently, a study of the contact electrification of micron-sized silica particles have also demonstrated that the surface charge density of the charged particles closely distributed around a constant value of 0.02 mC m-2 (that is, particles having a saturation or equilibrium charge), which is most possibly caused by the alternative mechanisms such as electron field emission and charge spreading at the particle contact site (Alois et al., 2017).

(iii) Line 342-344 in the manuscript has been modified as follows:

The proposed reasons for this are twofold: On one hand, the presence of adsorbed water could increase surface conductivity and particle-particle effective contact area, thus facilitating the ion or electron transfer (McCarty and Whitesides, 2008; Alois et al., 2018); On the other hand, OH⁻ ions in adsorbed surface water could also act as charge carrier (Gu et al., 2013; Lacks and Sankaran, 2011; McCarty and Whitesides, 2008).

References:

Alois, S., Merrison, J., Iversen, J. J., and Sesterhenn, J.: Contact electrification in

aerosolized monodispersed silica microspheres quantified using laser based velocimetry, J. Aerosol Sci., 106, 1-10, <u>https://doi.org/10.1016/j.jaerosci.2016.12.003</u>, 2017.

Alois, S., Merrison, J., Iversen, J. J., and Sesterhenn, J.: Quantifying the contact electrification of aerosolized insulating particles, Powder Technol., 332, 106-113, <u>https://doi.org/10.1016/j.powtec.2018.03.059</u>, 2018.

Comments 05: As the authors point out in laboratory studies it appears that the charge concentration (per surface area) is a more useful physical parameter than charge to mass ratio (μ), e.g. line is it possible to derive such a parameter from these measurements? Alternatively information of charge per dust particle might in this case also be valuable.

Response:

To the best of our knowledge, both surface charge density and charge-to-mass ratio are very important physical quantities for the electrification of granular materials. As the previous studies found that surface charge density played a key role in investigating particle charging mechanism (Alois et al., 2017; Lacks and Sankaran, 2011; Merrison, 2012). And charge-to-mass ratio is very critical for determining the effects of electrostatic forces on particle motion (transport), because the acceleration of dust particle due to electrostatic forces can be described as cE (where c is the charge-to-mass ratio and **E** is the electric field).

In this study, we cannot obtain the surface charge density and charge per particle because we only measured the mean space charge density and mass density (the information of the number of dust particles and real-time size distribution cannot be obtained).

Comments 06: It is of great interest that the observations presented here show a dependence upon RH especially as stated by the authors that the composition of the dust (soil) might imply a sensitivity to surface moisture (line 309). As the authors also point out some studies demonstrate dependence upon RH and others do not (line 337-347). Recent work has also shown that electrification can occur at extremely low RH

but that high RH may greatly enhance electrification for some materials. This appears to present a consistent picture (Alois 2018).

Response:

According to the reviewer's comments, we have cited the related recent work (i.e. Alois et al., 2018) in section 4.2 in the revised manuscript, as follows:

While water is not necessary for contact electrification (Baytekin et al., 2011a), a variety of studies indicated that such charge separation was strongly dependent on the RH (Esposito et al., 2016; McCarty and Whitesides, 2008; Xie and Han, 2012; Alois et al., 2018; Zhang et al., 2017). The proposed reasons for this are twofold: On one hand, the presence of adsorbed water could increase surface conductivity and particle-particle effective contact area, thus facilitating the ion or electron transfer (McCarty and Whitesides, 2008; Alois et al., 2018); On the other hand, OH– ions in adsorbed surface water could also act as charge carrier (Gu et al., 2013; Lacks and Sankaran, 2011; McCarty and Whitesides, 2008).