

## ***Interactive comment on “Direct observations of the atmospheric processing of Asian mineral dust” by R. C. Sullivan et al.***

**Anonymous Referee #2**

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This paper presents interesting results that give us insight into the mechanism of chemical ageing of mineral dust and helps to validate model predictions of time dependent mineral dust composition. Further evidence is presented for the accumulation of acidic pollutant gases by mineral dust during transport and the authors provide a reasonable explanation (titration of alkalinity followed by displacement of nitrate/chloride by sulphate) for observations that nitrate and chloride are found preferentially in the supermicron particles, whereas sulphate is found preferentially in the submicron particles. Further segregation of sulphate and nitrate is suggested to result from the greater SO<sub>x</sub> emission compared to NO<sub>x</sub> in Asia and interaction of dust with sulphate prior to nitrate/chloride. Ammonium was found only in dust particles with acid content and ammonium sulphate was found to be internally mixed with submicron dust aerosol, in

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contrast to previous interpretations. The authors also found that particle mineralogy had an important influence on e.g. sulphate versus nitrate uptake.

The article has novel results and hypotheses, is clearly written and well presented. The authors may wish to address the following comments:

General comment:

The authors show that mineral dust is chemically aged during transport from the source region to the RHB. They hypothesise that acidic trace gases such as  $\text{HNO}_3$  are the cause of this. A simple calculation using an approximated transport time and a rate expression (uptake coefficient etc.) for the uptake process would be useful to test this.

P4111, L19 Other laboratory data for uptake of  $\text{NO}_y$  to mineral dust exist: e.g. Vlasenko et al., (2006, Atmos. Chem. Phys. Disc), Hanisch and Crowley (2001, Phys. Chem. Chem. Phys).

P4115, L23 I've read this manuscript assuming that the analysis is conducted online. This statement appears to suggest that reference spectra were obtained from dust samples collected from source regions. Is this correct ?

P4116, L5 The word \*that\* appears twice

P4116, L23-24 State what the acronyms LAMMA, SIMS and LDI stand for.

P4121, L4 The rate of oxidation of  $\text{NO}_2$  on a dust surface is probably low. Can  $\text{NO}_2$  be considered a reactive gas when compared to e.g.  $\text{HNO}_3$  or  $\text{HCl}$  ? Are uptake coefficients for these species known ?

P4125, L20 There is no need to abbreviate ammonium nitrate to AN. Avoid the mixed use of chemical name, abbreviated chemical name and chemical formula.

P4126, L17 The implication of this sentence is that a single dust particle can support uptake of both acidic and basic species, prerequisite for the uptake of basic species being that acidic sites are available. The authors may note that uptake of acidic gases

may not only be restricted to reactive (alkaline) sites and that physisorption can also be important. Physisorbed acid gases will react with ammonium without consumption of alkalinity.

P4132, L3 Apart from Ooki and Uematsu, there are several laboratory measurements that compare the reactivities of SO<sub>2</sub>, NO<sub>2</sub>, and HNO<sub>3</sub> with mineral dust. These studies also show that HNO<sub>3</sub> is significantly more reactive than SO<sub>2</sub> or NO<sub>2</sub>.

P4139, L10 It is suggested that the presence of e.g. Fe enhances the oxidation rate of SO<sub>2</sub> to sulphate on dust surfaces, and that uptake to calcite particles is less efficient. Is this consistent with the observation of Usher et al (2002, J. Geophys. Res.) that SO<sub>2</sub> is (slightly) more efficiently taken up to CaCO<sub>3</sub> than Fe<sub>2</sub>O<sub>3</sub> ?

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 4109, 2006.

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