

Review of Lee and Kim, “Effect of the exclusion of crustal ions (Ca^{2+} , Mg^{2+} , and K^+) in estimating water content of $\text{PM}_{2.5}$ at polluted and clean areas”

While the subject of this paper falls within the scope of ACP, its lack of novel or important results and the extremely poor standard of writing means that it should not be accepted for publication in this journal.

This manuscript requires copy-editing or serious attention from a native English speaker to correct the grammar, spelling mistakes, and lack of clarity in many explanations. I had a lot of difficulty understanding the authors’ meaning in many sections as a result of the frequent typos and poor choices of vocabulary.

The authors use as motivation the possible reduction in dust emissions in NE Asia resulting from future increases in rainfall and/or standard of living. Their approach is to take historical observational datasets from Seoul and Gosan and use 24-h average measurements of inorganic $\text{PM}_{2.5}$ composition and associated gases as input for the SCAPE thermodynamics model. These observations contribute little beyond providing various sets of conditions that are consistent with two locations in Korea to use as model inputs, and the advantage of using these real atmospheric observations rather than arbitrarily-chosen values is not clear. Their results show that removing the crustal elements from the full suite of inorganic constituents tends to influence modelled aerosol water content, in one direction or another, as a result of changing the deliquescence RH and/or the remaining inorganic composition of the aerosol.

A serious weakness of the analysis is that there are no measurements of the actual water content of the measured particles, so that the model being used to predict possible changes in water content cannot be validated for the unperturbed state. The authors also do not address whether the model does a reasonable job of correctly predicting the gas-particle partitioning of the inorganic constituents, at least in Seoul where the use of denuders means that the measurements of partitioning may be reasonably accurate.

There are several reasons that the historical datasets selected by the authors are not ideal for this analysis:

- 1) The filter samples (and the SCAPE model) cannot account for variability in particle composition and one must assume that the particle population is internally mixed. Given the focus of this analysis on crustal elements, which are more common in super-micron particles, this is a major disadvantage. It is extremely unlikely that the NH_4^+ and much of the SO_4^{2-} is actually found associated with the crustal elements in the same individual particles. This means that many of the salts predicted and displayed in Figures 2 and 3 likely don’t exist in the real atmosphere being sampled.
- 2) There are no measurements of carbonate, which is likely quite important in samples where crustal elements are elevated and minimal acid displacement has occurred.
- 3) Over a 24-h sampling interval, the temperature and relative humidity are likely to have had significant variability. Using the daily average RH and applying it to the

daily average aerosol composition to estimate water content is almost certainly not appropriate.

The authors allude to this last point in the manuscript at the end of section 4.3, stating: *“Withstanding the fact that the aerosol water content is determined largely by relative humidity and that the relative humidity varies dramatically over the course of 24 h, it is hard to be seen that the result from this study represents the bulk composition at any one time during the day. Although it is a limitation of a time integrated sampling system, our result is valid for the purpose of predicting future trend change.”* Beyond the unsurprising ‘result’ that changing aerosol composition is likely to lead to a change in aerosol water content, I disagree that modelling of 24-h-averaged samples with no size-segregation can yield much quantitative information about the impact of crustal materials on aerosol composition and hygroscopicity.

It is not clear whether the data in Figure 1 represents the fraction based on mass or moles and this should be stated. The authors should show how closely charge balance was achieved in the historical data samples. This could help to explain the significance (or lack thereof) of carbonate. Also, there may be organic acids and bases that contribute to the charge balance and aerosol water content.

The authors have not explained how they remove the crustal ions in the model and maintain charge balance. What happens in the model – are the missing cations assumed to be replaced with H^+ or just removed from the system entirely? At any rate – does this really represent the ‘perturbation experiment’ articulated in the introduction, that is, a reduction in dust emissions? Wouldn’t that also reduce some other constituents at the same time?

Table 3 – why is KNO_3 the only salt that has a (s) subscript?

It is confusing that the authors choose different symbols for the data in Figures 2 and 3.