

Response to Anonymous Referee #1 comments

The authors present an analysis of submicron aerosol composition, CCN activity, and light scattering measurements made in the Eastern Mediterranean in 2012. They use the available data and modeling techniques to infer aerosol liquid water content and aerosol pH for the submicron aerosol fraction. Given the limitations of the dataset, specifically the absence of gas-phase NH₃ data for most of the measurement period, the analysis is consistent with the state of the art. Daily PM₁₀ filter composition analysis was also performed and this information was used to calculate LWC and pH for PM₁₀. I believe this manuscript is publishable in ACP after the following points are addressed directly in the revised manuscript.

Response: We thank the anonymous referee for the thoughtful review. Most of the issues raised were also concerns of the anonymous referee #3 therefore we have further elaborated on these points in the revised manuscript.

1) More discussion of the error introduced to the pH estimate by the absence of NH₃ (g) data is required. I will note that the "spot check" comparing the ISORROPIA predicted NH₃ (g) and the available measurements of NH₃ (g) mentioned at the end of section 2 is reassuring, but this discussion should be expanded and more technical information provided for the reader.

Response: To address the issue that is also raised by anonymous referee #3 (see also extensive reply to reviewer #3 general comments) a sensitivity study was performed and the more important findings will be clearly stated in the revised version of the manuscript. By adding different amounts of gas-phase ammonia to the initial results and computing ammonia's impact on predicted pH, we show that adding an amount of 5 $\mu\text{g m}^{-3}$ of gas-phase ammonia that is an extreme upper limit for ammonia concentrations in the studied region, pH values differ by a maximum of 1 unit (1.4 vs 2.4). Based on climatology data from the area (3-year study of Kouvarakis et al. 2001) gas phase ammonia ranges from 0.02 to 1 $\mu\text{g m}^{-3}$ with a mean and median value of 0.32 and 0.28 $\mu\text{g m}^{-3}$, respectively, which is even lower than the lowest added amount in the sensitivity study. Therefore the fact of neglecting the gas phase in the calculations has a difference of around 0.5 units in the pH (from 1.38 to 1.85 median values).

2) More information needs to be provided regarding the LWC and pH analysis for PM₁₀, including a frank assessment of the error bars on the pH and LWC calculations for PM₁₀. I did not see any of this mentioned in section 2, and PM₁₀ is mentioned only briefly at the beginning of section 3.4. The difference in available data and the level of analysis performed on the PM₁₀ vs. PM₁ needs to be discussed, especially since these datasets are discussed in conjunction in the later sections of the paper, and given the emphasis in the paper on bioavailability of mineral dust nutrients.

Response: We agree that the PM₁₀ results are subject to more uncertainty, as the inorganic semi-volatiles are not in equilibrium. With this said though, the liquid water does equilibrate rapidly and the ionic species therein – hence the calculations provided can be used as an indication of the coarse mode acidity with respect to the fine mode. This will be clearly stated in the revised version of the manuscript. Added information in the methodology section will include the differences in available data for the PM₁ and PM₁₀ fractions.

TABLE 1 -

The error bars on pH values listed here seem to be misleadingly small if the analysis itself introduces at least unit error to the pH estimates for submicron aerosol. How were they calculated for PM1 and PM10?

Response: We understand that the standard deviations (SD) of the pH estimates appear small and seem not to reflect the error introduced by the absence of gas phase ammonia data. Nevertheless, the variance of the values is expected to be different than the method bias. A clarification will be added in the text that apart from the standard deviation there is also the additional bias that is not expressed in the SD. Furthermore, the source/region data correspond to 7 or 9 days of selected data, representative of these sources/regions.

As Finokalia PM₁ aerosol contains a considerable amount of sulfates that does not vary significantly with day for the whole time period studied (June-November 2012), this is expected to limit the variance of the estimates. From long-term measurements of the aerosol -composition at the site, the relative contribution of the main PM₁ constituents, including ammonium, is quite consistent over the years (e.g., Mihalopoulos et al., 1997; Kouvarakis et al., 2001; Sciare et al., 2003; Koulouri et al., 2008, Bougiatioti et al., 2009; 2011; 2013). A fluctuation of 10-20% in sulfate and/or ammonium concentrations is not expected to be reflected in a pH change, given the logarithmic scale of the property. On the other hand, PM10 pH estimates express the bulk acidity that covers a wider range.

FIGURES -

The time series presented in Figures 1 and 2 are not easily legible in the current format, these figures should be redesigned in order to have more meaning for the reader.

Response: Both figures are now converted to a stacked format on the same scale and are easier for the reader to follow.