

The manuscript titled “Spatial and temporal distribution of fine aerosol acidity in the Eastern Mediterranean” by Neroladaki et al. characterizes aerosol acidity in Greece by comparing and contrasting measurements among six geographic sites and between summer and winter time periods. Using an aerosol thermodynamics model, the study finds that pH is lower during the summer than winter due mostly to sulfate variability, that organics do not always increase pH when aerosol pH is higher during the winter, that aerosol pH is more sensitive to NH_3 during the summer since pH is lower, and that NH_3 deposition is fast while HNO_3 deposition depends on environmental conditions. The study provides interesting insights and has potential to increase understanding on aerosol acidity, however, there are some areas that are unclear or lack necessary supporting information that should be addressed prior to publication. Following these revisions, I recommend publication.

Specific Comments

Line 112: Remove “, trend”

Line 185 (and throughout the manuscript): The text jumps back and forth between the full spelling of chemical species and their abbreviations. In one sentence starting on Line 184, “ NH_3 ” is first used then “ammonia” is used several words later. For clarity and consistency of the manuscript, it is recommended to stick to only the abbreviations or only the full spellings.

Line 199: add “are” before “1.24...”

Section 2.2: For the gas-phase measurements, the manuscript utilizes a combination of simultaneous in-situ measurements at its site, simultaneous in-situ measurements at a neighboring site, past in-situ measurements at its site, past in-situ measurements at a neighboring site, and satellite measurements at its site that are averaged over 1 year at some sites and over 14 years at another. While the text at present explains the various measurements, it is very confusing and difficult to keep track of this information (which is relevant to understanding the results and impact of the study). It would be beneficial to create a table that summarizes the type of gas-phase measurements (satellite vs. in-situ, simultaneous vs. past, same site or neighboring site) for each of the 6 sites. This would allow for a simplified comparison of the measurements used and can serve as quick reference for the reader while going through the results section. Such a table may be included in the supplement and referenced from the main text.

Section 2.2: It is not clearly stated in the methods that summer measurements are not available at all 6 sites. Table 1 is the first (indirect) mention that PTR, XAN, and LAP only have winter measurements. The time frame of measurements at each site should be explicitly stated in the methods. Additionally, it is suggested that measurements are during the summer of 2019 and winter of 2019-2020. Later in the results section, it is mentioned that only January 2020 measurements are used. January 2020 would be winter 2020 not winter 2019-2020. Please clarify in the methods section the specific measurement times, and use only one same term (either winter 2020 or January 2020) throughout the manuscript for consistency.

Line 205: Why is the median used for HNO_3 and HCl at FKL while the mean was used for HNO_3 at all other sites (and the mean was also used for satellite measurements)? Please justify the one use of the median while all other cases utilize the mean.

Section 3.2.4: Are the differences in pH between summer and winter statistically significant (e.g. by a two-sample t-test)?

Section 3.3: A total of 9 parameters were tested at IOA and THI but only 4 parameters were tested at FKL. This leaves RH, TNO₃, Na, Ca, and OA missing at FKL. Why were these not run for FKL? Based on the averages listed in Table 1 these measurements are available and so there is not an apparent reason for why they were not considered at FKL. Please either add the 5 missing parameters for FKL or justify why they were not considered.

Section 3.4: What are the implications of the results of this sensitivity analysis specifically to the results of this manuscript? It seems that this sensitivity analysis is particularly relevant to the results at the IOA site since satellite measurements were used during the summer but in-situ measurements from PTR were used during the winter. The results of the change in aerosol pH between summer and winter (Section 3.2.4, Figure 6) then could possibly be muddled by differences in gas-phase measurements demonstrated in Section 3.4. Please discuss.

Figure 5a: The plot lines and axis labels match in color but the axis ticks are mis-matched in color.

Figure 6, 8: The axis labels are too small to be legible. Please make them larger or consider using different abbreviations.