

We would like to thank the reviewer for the constructive comments and feedback on our manuscript. We considered all comments and have revised our manuscript accordingly. We provide below a point by point response to the comments. Reviewer comments are shown in *italic* and our responses follow.

Reviewer 1

Comment 1:

Line 112: Remove “,trend”

Response:

Removed.

Comment 2:

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, “NH₃” 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.

Response:

In the revised manuscript we first provide the full name and abbreviation of the chemical species and then only the abbreviation is used.

Comment 3:

Line 199: add “are” before “1.24...”

Response:

Added.

Comment 4:

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.

Response:

We thank the reviewer for this comment, which helped increase the clarity of our study. As suggested by the reviewer, we have added a summary table that provides the type of gas-phase measurements for each of the 6 sites. It is now included in the supplement as Table S2.

Comment 5:

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.

Response:

In the revised manuscript, we now clarify that FKL, THI and IOA are the only sites with available summertime measurements. The campaigns took place in summer 2019 and winter 2019-2020. However, the timing of the measurements differs from site to site. Therefore, in order to be able to compare the aerosol pH among the sites for winter we only used January 2020 data, except in Section 3.2.4. In that section, where we investigated the seasonality of aerosol pH in the 3 sites, we used all available days with measurements in each site in order to include as many data points as possible, since we did not make a straight comparison among the sites and rather we investigate the seasonality in each site separately. We now use “January 2020” when only data from this month are used. Similarly, to properly compare the three sites in the summer, we used the common days with available measurements in July and August. Clarification is made in the revised version of the manuscript.

For winter:

“Due to the lack of data in some of the sites during the winter 2019-2020, the results of only January 2020 were used in order to be able to compare the aerosol pH results among the sites. The entirety of the aerosol pH results for all sites during winter 2019-2020 can be found in Fig. S4.”

For summer:

“Overall, comparing the summertime (July and August) aerosol pH levels at the three sites (Fig. 3c and S5), a uniformity can be observed with high aerosol acidity being the case on most of the days, dropping even below 0 at IOA as a result of increased temperature and sulfate levels and reduced aerosol water.”

Comment 6:

Line 205: Why is the median used for HNO₃ and HCl at FKL while the mean was used for HNO₃ 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.

Response:

Indeed we should have used the mean concentrations of the gases in order to be consistent. However, we recalculated the aerosol pH and water using the mean values and found that there is no significant difference between using median values. The aerosol pH and water were not statistically different from the ones calculated with the mean values with a confidence level of 95% (student two sample t-test and Mann–Whitney U test). Below we provide the results using both mean and median concentrations. The winter and summer results are in parenthesis; (w) and (s) respectively.

With median values: Aerosol pH → 3.25 ± 0.37 (w), 2.08 ± 0.37 (s)
Aerosol water → 6.85 ± 3.65 (w), 5.89 ± 3.05 (s)

With mean values: Aerosol pH → 3.22 ± 0.34 (w), 2.08 ± 0.45 (s)
Aerosol water → 7.10 ± 3.81 (w), 5.41 ± 3.24 (s)

Comment 7:

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

Response:

The differences in pH between summer and winter are indeed statistically significant with confidence level 95% in all 3 sites (FKL, THI, IOA). We use both a student two sample t-test and Mann–Whitney U test.

Comment 8:

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.

Response:

We used only 4 parameters in FKL instead of 9 as in IOA due to their insignificant effect on seasonality. The same effect occurred in THI, though. In order to be consistent, we repeated the sensitivity tests in FKL and in THI considering all 9 parameters. The simulations are now consistently named among the sites (S₁ is always the simulation when temperature is tested, S₂ when relative humidity is tested, etc.). The corresponding panels in Fig. 6 were replotted. We have modified the manuscript in section 3.2.4 and Tables S5 and S7 in the supplement accordingly.

Comment 9:

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.

Response:

Indeed, the differences in the input data used motivated us to perform this sensitivity analysis. The results of the sensitivity analysis are now further discussed for IOA following the reviewer comment.

“According to this sensitivity analysis, the uncertainty in the gas phase NH₃ could explain about half of the seasonal difference in aerosol pH that was presented and discussed in section 3.2.4 about the factors affecting the seasonality of pH. The uncertainty in gas phase NH₃ could explain a larger fraction of the seasonal difference in aerosol pH in IOA than in FKL and THI, which is expected since at these two sites aerosol pH seasonality was predominately driven by sulfates. Our results show a factor of 2 uncertainty in NH₃ lead to an average pH difference of 0.25 units that has minor impact on our findings discussed below.”

Comment 10:

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

Response:

We thank the reviewer for pointing out this mismatch that is now corrected in the revised figure.

Comment 11:

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

Response:

Indeed, the figures were not easily legible. We revised those figures and increased the size of the axis labels.