Ditto et al. present a unique dataset on the molecular composition of nitrogen-containing organic compounds in aerosols and the gas-phase. Overall, the methods appear sound, although it requires reading some of their previous papers to establish this. The results presented are interesting and relevant to our understanding of organic aerosol chemical composition. The main concern I have with the manuscript is the lack of engagement with other studies on organic N in rain and aerosols, as well as other high-resolution studies of organic nitrogen chemical composition. The authors conclusions are similar to what some other studies have found, and it would lend additional credibility to this study to engage with the literature more thoroughly.

Title – given the small sample set it seems an over-reach to title the paper a “seasonal analysis.” It is more akin to a case study with summer and winter cases. There is not enough information to truly explore seasonality.

Introduction/Discussion – It would be useful to introduce readers to similar studies. This could be done in the introduction or in the discussion. For example, the following seem particularly pertinent to this study and find similar results (with respect to the dominance of reduced N compounds): (Altieri et al., 2009; Podgorski et al., 2012; Spranger et al., 2019; Wozniak et al., 2014).

Other related studies include: (LeClair et al., 2012; Mace, Artaxo, et al., 2003; Mace, Kubilay, et al., 2003; Romonosky et al., 2015; G. Zhang et al., 2020; Q. Zhang et al., 2002).
This is obviously not an exhaustive list but highlights that the authors have not really gone beyond comparing the results here to their own previous work. This needs to be rectified in order for the manuscript to be publishable in a journal such as ACP.

Full citations:


Methods:

The sampling durations are too short and sporadic to be seasonally representative.

Typo page 6 line 130 “loses” should be “losses”.

Page 6 paragraph line 133 – what type of air sampler and filter holders were used? What size aerosols were selected for – or was it bulk? More detail is needed.

More detail is needed on the elemental formula assignments and the handling of blanks. I had to read the entire previous Ditto 2018 and 2020 papers to feel fully satisfied that it was done properly.

Page 7 line 146 – it’s not clear what the choice of “examined” means here, perhaps “identified” is more appropriate? If this is not correct, then the sentence needs to be re-written for clarity.

Page 10 line 212 – more information is needed on the typical time scale of hydrolysis for the expected compounds of interest including appropriate references.
Page 11 line 237 – more information is needed on the make, model, pump, and size cut-offs of the aerosol sampling system.

Page 11 line 243 – why only 48-hour back trajectories? This seems short given the lifetime of PM$_{2.5}$ and some of the gas-phase organics. Some justification is required.

Page 11 line 246 – Similarly, why was a height of 50m chosen for a ground based sampler? I am assuming it is to get above the errors associated with surface topography, but a description of the choice is required.

Results and Discussion:

3.2.1 – significantly more discussion is needed to explain the determination of IVOC/ULVOC/SVOC/LVOC/ELVOC. The only information presented is in the caption of figure 2 and it is insufficient. The reader must be able to judge if they agree with the classifications.

Page 13 line 282 – what does the p-value indicate? Is this a t-test to determine if the ozone concentrations are significantly different? Similar questions page 14 lines 290

Page 15 lines 317-319 – references are needed here or a more detailed discussion on why aqueous-phase processing leads to the stated shifts.

Page 15 line 328 – in this case it seems impossible that the stated p-value is from a t-test suggesting those are different values. Explanation is required.

Page 16 line 329 – why is in-cloud processing not considered as a contributor to aqueous-phase chemistry throughout the discussion?

Figure 2 – Is ion abundance intended to be a proxy for concentration? How was the average volatility determined? How were the volatility classifications assigned (more is needed in the text than the simple statement in the caption). What is the log(C0) and how
was it determined? The text on lines 358-363 needs to be in the manuscript and not the figure caption. Furthermore the impact of using the same temperature for the summer and winter cases will be significant such that it’s not clear what this comparison is actually showing given that the results for winter will be completely different.

3.2.3 – this is not a comparison to other sites, it is a comparison to the authors previous work. Even if the methods are slightly different, surely some other studies can be compared here?

Page 19 line 388 – first mention that these are PM10 samples

Page 19 lines 389-393 – Some data or figure needs to be presented to justify the statement here.

Figure 3 – Given that it is known that ionization efficiencies vary for the different groups of compounds, it seems to make more sense to use the number of compounds not weighted by compound abundance. The weighting is related to both the concentration and ionization efficiency in an unknown manner so it is not clear what the weighting actually means for the results.

Page 21 paragraph starting on line 424 – given the discussion of possibilities, it would be useful if the authors proposed what they think is the most likely driver in winter. For example, it is probably not the marine source given that surface ocean productivity should be at a minimum in winter. But what is most likely?

The reference on page 22 line 460 seems out of place – the sentence reads as if these assignments are for the current dataset but then refers to previous work. This needs to be clarified. Were some of these samples published in the previous study?

Page 25 line 519-520 – this seems indicative of a marine signal?

Page 33/31 lines 632-633 – but this was significantly less so in winter, correct? So what is the connection?