

The authors have made substantial improvements addressing the referees' concerns, by presenting additional supporting plots and explanations, putting the measurements more in context of other studies, and reframing the implications and conclusions to better reflect the evidence presented. However, there still appear to be some explanations and conclusions drawn that are not well supported – and therefore should not be published in current form. Details are below along with a few other suggestions/questions.

Section 3.4 and Fig. 6 on model/measurement comparisons:

It is stated “The concentrations of BC and LO-OOA generally decreased slowly after sunset till sunrise due to the combined effect of both the planetary boundary layer variation and traffic emissions while particulate organic nitrates showed a different trend with two clear growth processes (19:00-22:00 and 3:00-6:00) at night, suggesting their unique sources. In contrast, the production potentials of the six notable VOCs with NO₃ had two roughly similar increases at the same periods as those of particulate organic nitrates, which supported the key role of NO₃+VOCs reactions for nighttime organic nitrate formation.”

How do PBL and traffic emissions explain the steady decline of BC and LO-OOA? More clarification is needed on why this would be the case and how it is known. Generally, statements like this seem to be made a bit haphazardly throughout the manuscript – i.e. strongly stating an explanation for something that is likely a complex process, while providing little explanation/evidence. Loose language/science like this is detrimental to the scientific literature.

More importantly, the statement that the nocturnal cycles of the PP and organic nitrate match is simply not supported by the data! It seems to rather describe what the authors hoped to see in the data based on the modeling. The PP for the dominant SOA-forming compounds shows a 2-4 fold increase at 21-1 hrs while the organic nitrates show subtle decreases. Thus, the observed aerosol data does not seem to support any connection between modeled sources and measurements based on what is shown. The authors should revise the discussion accordingly to accurately reflect what can be supported with the data. As is, it seems to suggest that either the modeling framework does not represent the dominant chemistry producing organic nitrates and SOA or there are other factors at play that make the effects difficult to observe. Perhaps there are other more important anthropogenic precursors, that were not measured, that build up during the nighttime in the lower nocturnal BL and react with NO₃ or O₃? That would be consistent with the stronger correlations with NO₂ than with the VOCs show in Fig. S11.

As I suggested in my initial review, the subtle features in the organic nitrates may be just driven by averaging artifacts and further investigation of time series or correlations may shed some light on the connections (or lack thereof). For example, during nights when the PP is particularly high, are the organic nitrates and LO-OOA correspondingly elevated? How do the correlations of LO-OOA and organic nitrates vs calculated PP look? Or better yet, versus computed particle organic nitrate or SOA production. Without further compelling observational evidence, any conclusions about the importance of particular VOCs+NO₃ sources should be strictly framed as theoretical, based on the modeling of potential sources throughout the paper. In fact, in that case, the possibility that other anthropogenic VOC may be driving the nighttime organic nitrate production should be considered and discussed.

Also, if the authors choose to keep Fig. 6, it would be very useful to include the nocturnal cycle of the calculated SOA and organic nitrate formation rate since that is more directly related to the particle-phase measurements being discussed.

Sect. 3.3 on chemically-resolved size distributions:

This is an interesting addition since the original manuscript. To my knowledge high-resolution nitrate and/or NO⁺ and NO₂⁺ has not been reported before in the published literature. The authors should confirm that the NO⁺/NO₂⁺ ratio was computed using high-resolution PToF analysis (since UMR m/z 30/46 would be much less meaningful due to CH₂O_x⁺ interferences). Therefore, can the authors please show size distributions of the NO⁺ and NO₂⁺ signals for the different seasons (in SI)? This would help to understand how well the HR PToF worked in separating those two ions and provide a helpful example for future analyses using AMS HR-PToF data.

Additionally, if this is indeed HR-PToF, I would strongly encourage that the authors compute the size distributions of organic and inorganic nitrate separately (using the NO_x⁺ ratio equations and the total nitrate, NO⁺ and NO₂⁺ size distributions) and show them in Figure 4. That would allow more direct and intuitive inspection of where the different modes reside (particularly for the non-AMS aficionado). And relatedly, showing that organic nitrates size distributions are indeed significantly different from the OA distributions is critical to support the interpretation in this section that the size-dependence of the NO_x ratio indicates fresher SOA sources.

Section 3.5 on comparison to other studies:

It is stated “Note that, particulate organic nitrates constituted the major part (86-100%) of total nitrates in the atmosphere scarce of NO_x (in Centreville and Woodland Park), suggesting that NO_x was very quickly consumed to form particulate organic nitrates and thus the formation of particulate organic nitrates should be NO_x-limited.”

This is wild speculation and almost certainly false. The organic nitrate concentrations at those sites were much lower than NO_x and simply the fact that organic nitrate >> inorganic nitrate does not provide a direct line of reasoning to what the major losses of NO_x are. Lower NO_x at those sites was likely due to the fact they were removed from strong urban emissions. Moreover, the use of “NO_x-limited” here and throughout the paper is loosely and ambiguously invoked. With lower NO_x concentrations, NO₃ radical production may decrease, however the BVOC will still most certainly instead react with O₃ or OH and form organic nitrates and SOA. In this section and throughout the authors need to clarify what is meant by NO_x-limited – including discussion of other literature for context. As is, all discussions of “NO_x-limited” is ambiguous and underdeveloped. I think for the analysis of this particular study being analyzed, the authors mean that it is driven by NO₃ chemistry. But that is not the same as “NO_x-limited”.

Table 2 and caption and manuscript text: variability with FPEAK is not a metric of “uncertainty”.

Bootstrapping is typically used as an estimate of uncertainties. “Uncertainties” in this context should be replaced with something like “variability”. Also, it is not clear what the number in the table reported is. Is that the standard deviation of all values with different FPEAK, or the max difference from the FPEAK=0 solution? Also it should be written as FPEAK, not fpeak.

Figure 6: “alpha” is misspelled.

Table S3. Alpha and beta Pinene are misspelled.

General: There are a quite a few grammatical mistakes, particularly in the new text that was added.