

Interactive comment on “On the contribution of nocturnal heterogeneous reactive nitrogen chemistry to particulate matter formation during wintertime pollution events in Northern Utah” by Erin E. McDuffie et al.

Anonymous Referee #2

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Overall, I find this to be a very nice paper that addresses the important issue of particulate nitrate formation mechanisms in polluted valley regions. It is well written, with very nice figures, and a generally thorough analysis, although limited by non-consideration of daytime formation and most loss processes. I suggest that it be accepted once the authors address the issues below.

General Comments:

In the box model, does the loss rate of N_2O_5 have any impact on the O_3 and NO_2 ? If,

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hypothetically, N₂O₅ loss is set to zero, then the O₃ and NO₂ would evolve differently than if the loss is fast, correct? The model, by separating the O₃ and NO₂ optimization and the N₂O₅ and ClNO₂ optimization seems to neglect this. Is this a concern? Is the robustness tested by then using the derived gamma_N₂O₅ and ClNO₂ yields to ensure that O₃ and NO₂ profiles are unchanged?

Fig. 6: For the night/day P(NO₃) calculations, in panel a I find the legend descriptions to be a little confusing. Specifically, the meaning of “~ RL at night” confuses that this is daytime.

Section 3.3.1: It seems like it would be a good idea to add a subscript (or some other indicator) of “max” to the P(NO₃) values to make sure it is clear that these are the maximum. This is especially important for e.g. Fig. 6, where that context is not readily apparent.

Fig. 6b: Is there a reason that the number of points at a given altitude is so different between NO₂, O₃, and PNO₃ (and PM₁)? Does this have to do with estimation of the surface area and differences in averaging time?

P11/L33: Table S4 doesn't seem to address how uncertainties impact the ClNO₂ yield, only net nitrate production. The paragraph, as written, sort of makes it seem like there is an implication of the ClNO₂ yield being relatively insensitive to other uncertainties.

P12/Hygroscopic growth: Related, but not directly addressed, is that the time history of RH may matter. The authors use the RH at the point of measurement and, somehow, extrapolate this back in time in the model to give a minute-by-minute perspective on particle surface area. Historical fluctuations in RH could influence the point observations. Is there an attempt to account for variations in RH with time of day? Similarly, the authors seem to use a fixed dry SA, based on the intercept point. But if aerosol growth is occurring then the SA would evolve over time. What sort of uncertainty does this simplification bring in?

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P12/L13: It should also be noted that the steady state approximation requires the surface area and temperature, not just NO₂, O₃, and N₂O₅.

P13/L33: It would be good if the authors commented here on the substantial variability in the derived values and, perhaps, the seeming bimodality (with a high production mode and a lower production mode).

P14: The discussion of dilution/entrainment could be enhanced. The Womak paper was just published on 4/8, making it available. It seems that the authors here are arguing for an entrainment rate that is largely independent of time of day, except for the issue of changing height of the mixed layer. However, one might expect the entrainment rate to differ notably between the daytime and nighttime. It is unclear whether the authors are applying a daytime entrainment rate to the nighttime or really, in general, how entrainment is being accounted for. The origin of the 40% scaling factor is somewhat mysterious as well.

P15/L3: The difference between the L and M cases seems negligible, as is evident from Fig. 10. Why include both of these when they are so similar?

P15/L4: Should the transition from nighttime to daytime also be accounted for? In other words, if most nitrate is formed aloft (as suggested) and the air in the RL is entrained to the surface starting at sunrise, then the nitrate in the RL will be distributed throughout the daytime mixed layer. Dilution from exchange with the FT will occur on top of this. However, that would also require accounting for the nitrate in the surface layer initially.

P15/L9: I have some difficulty with the framing here. The authors start by saying that nocturnal chemistry and largely explain the nitrate accumulation at the surface. But then they go on to say that Womack (who looked at the same events) concluded that photochemical production is quite important too. Notably, while Pusede et al. (2016) implicated nocturnal nitrate production as very important, they did not discount daytime production to the extent suggested here and Prabhakar et al. (2017), building on Pusede et al. (2016), concluded that daytime production plays an important role. I

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suggest that the authors consider revising the first sentence and how they frame the discussion here. They have only considered nighttime formation and thus their ultimate conclusions regarding the contributions of daytime processes are limited.

Minor points:

A minor grammar question: should it be “in the SLV” or “in SLV”. I would have thought the former, as the SLV is not a city but a region (like the western US, which the authors refer to with a “the”).

P13/L5: “empirically-based” should just be “empirical.”

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