

Review of Observational assessment of the role of nocturnal residual-layer chemistry in determining daytime surface particulate nitrate concentrations by Prabhakar et al.

The authors study the role of nocturnal chemistry and transport processes in determining surface PM_{2.5} levels and provide evidence for the morning transport of NO₃⁻(p) from the upper layers to the surface. This is a comprehensive study that includes an analysis of combined airborne and ground observations, and quantitative estimates of important parameters such as mixed layer height heights, loss processes of NO₃⁻, and N₂O₅ uptake coefficients. I highly recommend the publication of this manuscript in ACP after minor revisions.

1. The authors suggest in Page 8 line 25 “.. *the expected local nocturnal chemical production of nitrate in the RL should exhibit relatively minor vertical variation (due to variations in temperature and RH) (Figure S5). In other words, without loss or dilution processes, it is expected that the NO₃⁻(p) concentration would increase to a similar extent at all RL altitudes.*” and attribute the observed vertical distribution of NO₃⁻(p), which shows enhancements ~ 100 – 150 m agl at ~ 9:30 AM, to the differential horizontal advection in the RL. I am not 100% convinced that the horizontal differential advection is the main driver of the observed morning profiles of NO₃⁻(p). Figure 3a does NOT show maximum wind speed at ~ 250 m, which is also the altitude at which morning NO₃⁻(p) is minimum. Figure 3 suggests to me the strongest winds appear to be at 150 m very frequently, whereas on average they (b2 and b3) are basically the same through a deep layer and very light winds (~1.5 m/s). This is consistent with the idea that the horizontal transport aloft is responsible for the valley-wide distribution of pollutants/precursor gases, but it does not necessarily suggest its direct role in the vertical distribution of NO₃⁻(p). Also the average vertical profiles shown in Figure S3 and Figure 2A look very similar to the average nocturnal profiles of NO₂, O₃ and N₂O₅ reported by Brown et al. ¹, indicating the altitude dependent chemical composition due to stable conditions could account for the observed NO₃⁻(p) profile if heterogeneous uptake of N₂O₅ is the key source of NO₃⁻(p). Consider re-phrasing some of the statements.
2. The authors assume morning chemistry due to ClNO₂ and/or HONO is negligible even though the vertical profiling takes place 9:30 AM, after ~1 – 2 after sunrise and Brown et al. ² reports enhancements in NO₃⁻(p) during the night and early morning. More justification or comment on morning chemistry is needed.
In the same vein, the authors derive [OH] by using peak [OH] of 1x10⁶ molecules/cm³ at noon, a relatively large number under wintertime conditions (although conditions in California might be different than other areas with typical winter season) and by scaling it with the observed solar radiation and estimate the photochemical contribution to be large, ~ 60% at 3 PM. Discussion on the effects of snow cover on the ground, cloud cover and fog during these episodes will be helpful since typical cold pool conditions often include those, which in turn limit the photochemical production.
In general, more extended discussion contrasting pollution episodes in other areas will improve the overall quality of this manuscript.

3. Transport out or entrainment from free troposphere in the afternoon leads to the loss of $PM_{2.5}$, but it also introduces O_3 and dilutes NO_x^3 , impacting the P_{NO_3} formation at night. What about the later effects? Figure S3c indicates significant differences in O_3 above and below the inversion cap during this episode.
4. Page 13 line 17: "The peak and turnover in surface-level NO_3 -(p) occurs when higher RL layers, where $[NO_3$ -(p)]_{RL} < $[NO_3$ -(p)]_{ML}, are entrained.". Shouldn't it be " $[NO_3$ -(p)]_{RL} > $[NO_3$ -(p)]_{ML}"?
5. Using increase rate in 24-h $PM_{2.5}$ running average and % nitrate contribution might be more suitable for estimating the nocturnal nitrate production as it already take into consideration the transport term.

1. Brown, S. S.; Dubé, W. P.; Osthoff, H. D.; Wolfe, D. E.; Angevine, W. M.; Ravishankara, A. R., High resolution vertical distributions of NO_3 and N_2O_5 through the nocturnal boundary layer. *Atmos. Chem. Phys.* **2007**, *7* (1), 139-149. DOI 10.5194/acp-7-139-2007.
2. Brown, S. G.; Roberts, P. T.; McCarthy, M. C.; Lurmann, F. W.; Hyslop, N. P., Wintertime vertical variations in particulate matter (PM) and precursor concentrations in the San Joaquin Valley during the California Regional Coarse PM/Fine PM Air Quality Study. *J. Air Waste Manage. Assoc.* **2006**, *56* (9), 1267-1277. DOI 10.1080/10473289.2006.10464583.
3. Baasandorj, M.; Hoch, S. W.; Bares, R.; Lin, J. C.; Brown, S. S.; Millet, D. B.; Martin, R.; Kelly, K.; Zarzana, K. J.; Whiteman, C. D.; Dube, W. P.; Tonnesen, G.; Jaramillo, I. C.; Sohl, J., Coupling between Chemical and Meteorological Processes under Persistent Cold-Air Pool Conditions: Evolution of Wintertime $PM_{2.5}$ Pollution Events and N_2O_5 Observations in Utah's Salt Lake Valley. *Environ Sci Technol* **2017**, DOI 10.1021/acs.est.6b06603.