

We thank the referee for his/her time and commentary, which have improved our manuscript.

Best,

Sally and Ron

*The authors argue with past field observations that secondary production is the likely driver for ambient ammonium nitrate concentrations and report expected reductions in daily averaged PM<sub>2.5</sub> in the San Joaquin Valley for future reductions in NO<sub>x</sub>. The impact of NO<sub>x</sub> reduction on the nighttime production mechanism for ammonium nitrate formation is found to be more important in initial stages of reduction. The authors conclude this by building an observation-based model that considers net rates of ammonium nitrate production from diurnally-varying gas-phase and heterogeneous reaction pathways, loss by rapid dry deposition, and boundary layer meteorology. There are many parameters estimated for the model, but the assumptions are generally well documented. This approach provides an alternative to making such predictions by air quality models, which face difficulties on account of uncertain meteorology and emissions in this region. The framework presented in this manuscript is well grounded but some of some additional explanation or caveats can be introduced. The manuscript is valuable to the atmospheric chemistry community from a practical perspective, and is suitable for publication in Atmospheric Chemistry and Physics after addressing the following comments:*

*The PM<sub>2.5</sub> response to NO<sub>x</sub> reductions and corresponding exceedances of the 24-hour PM<sub>2.5</sub> standard are presented without enough qualification of the role of NO<sub>x</sub> on secondary organic aerosol formation. There is discussion of the NO<sub>3</sub> radical reaction with organic species (p. 27095) and requirements on the control of organic aerosol mass (p. 27102), but reduction in the RO<sub>2</sub> + NO reaction should lead to increase in SOA (e.g., Presto 2005). Given that the PM is mostly ammonium nitrate and organic matter in this region, this seems to be a very important point to make in the manuscript.*

*Presto, A. A.; Hartz, K. E. H. & Donahue, N. M. (2005): Secondary organic aerosol production from terpene ozonolysis. 2. Effect of NO<sub>x</sub> concentration, Environmental Science Technology, 39, 7046-7054, doi:10.1021/es050400s.*

We agree that NO<sub>x</sub> reductions will impact SOA; however, quantifying the impact of NO<sub>x</sub> reductions on SOA, or even speculating on the sign of the impact in the wintertime SJV, is beyond the scope of our work. We have added the text below stating that NO<sub>x</sub> will impact SOA and explaining that we have not included it in our calculation of the exceedance change:

Page 17, lines 3–16: “We have not quantified, but do expect, future NO<sub>x</sub> reductions to impact the portion of organic aerosol mass that is secondary (SOA). In the laboratory, it has consistently been observed that NO<sub>x</sub> concentrations, relative to gas-phase organic compounds, influence the molecular identity and volatility of oxidation products such that SOA yields are higher at low NO<sub>x</sub> and suppressed at high NO<sub>x</sub> (e.g., Presto et al., 2005; Ng et al., 2007; Kroll and Seinfeld, 2008; Chan et al., 2010). Recent summertime field measurements of aerosol-phase

RONO<sub>2</sub> in Bakersfield (Rollins et al., 2012) and at a forested field site in Colorado (Fry et al., 2013) found that NO<sub>3</sub> radical-initiated SOA formation correlated with NO<sub>3</sub> production and was proportional to NO<sub>x</sub> at low to moderate NO<sub>x</sub> levels. In Fresno during DISCOVER-AQ, SOA constituted 40% of the organic fraction of PM<sub>1</sub>, or 22% of total PM<sub>1</sub> mass (Young et al., 2015). Reductions in NO<sub>x</sub> as large as 50% to 75% are expected to influence this portion of the aerosol mass, and likely in a way that affects the frequency of exceedances in the SJV; however, the magnitude and sign of the impact are beyond the scope of this work.”

*Regarding the use of ISORROPIA II, why were the ammonia concentrations set to 1.1 times gas-phase nitric acid concentration (p. 27098)? Walker et al. (2012) suggests that many parts of California are ammonia-limited (including parts of the San Joaquin Valley). I suspect Figures 1 and 2 suggest otherwise for the studied locations, but this may be worth addressing.*

*Walker, J. M.; Philip, S.; Martin, R. V. Seinfeld, J. H. Simulation of nitrate, sulfate, and ammonium aerosols over the United States, Atmospheric Chemistry and Physics, 2012, 12, 11213-11227, doi:10.5194/acp-12-11213-2012.*

Figs. 1 and 2 imply NH<sub>4</sub>NO<sub>3</sub> is nitrate limited and we use this result to guide our decision to force NH<sub>3</sub> to be in excess. We modified the text to make this point clear:

Page 6, lines 23–28: “The key idea is that present day NO<sub>3</sub><sup>−</sup> concentrations on weekdays are equal to what were seen on weekends a decade ago, i.e. the NO<sub>2</sub> dependence of NO<sub>3</sub><sup>−</sup> has been unchanged with time. This suggests that in the wintertime average, the only source of NO<sub>3</sub><sup>−</sup> in the atmosphere has been oxidation of NO<sub>2</sub> and that NH<sub>4</sub>NO<sub>3</sub> production has been nitrate rather than ammonium limited. Agreement of NO<sub>3</sub><sup>−</sup> in different years at identical NO<sub>2</sub> implies that there has been little change over time in the chemical mechanism producing NO<sub>3</sub><sup>−</sup>, and hence NH<sub>4</sub>NO<sub>3</sub>.”

Page 7, lines 22–24: “We infer from Figs. 2 and 3 that the oxidation of locally emitted NO<sub>x</sub> is the single largest term affecting the production of NH<sub>4</sub>NO<sub>3</sub>, that NH<sub>3</sub> is in excess, and that transport and mixing are too slow to fully homogenize the aerosol throughout the wintertime SJV.”

Page 12, lines 9–12: “ISORROPIA II was initialized as [NO<sub>3</sub><sup>−</sup> + HNO<sub>3</sub>] = [NO<sub>3</sub><sup>−</sup>]<sub>AMS</sub> and [NH<sub>4</sub><sup>+</sup> + NH<sub>3</sub>] = [NH<sub>4</sub><sup>+</sup>]<sub>AMS</sub>. Calculated HNO<sub>3(g)</sub> was added back to [NO<sub>3</sub><sup>−</sup> + HNO<sub>3</sub>], while NH<sub>3(g)</sub> was added as 1.1 HNO<sub>3(g)</sub> (by mole) to ensure NH<sub>3</sub> was in excess because NH<sub>4</sub>NO<sub>3</sub> was shown to be NO<sub>3</sub><sup>−</sup>-limited (Figs. 2 and 3).”

*Data from various size cuts (submicron, PM<sub>2.5</sub>, PM<sub>3</sub>, PM<sub>10</sub>, etc.) are used throughout the work and not always clarified when referring to concentrations.*

We have added the size threshold of each observation in all figure captions and done our best to include this information in the main text at each instance where unclear.

*p. 27104 line 24: “If we assume ambient conditions are driving [...], we can estimate...” → This is a strangely worded statement.*

We have modified the sentence as follows:

Page 20, lines 21–22: “If we assume ambient conditions, as opposed to conditions internal to the instrument, drive the equilibrium...”

*Figure 5 caption. “Time follows the NO<sub>2</sub> trend.” is also worded strangely.*

We have modified the caption as follows:

**“Fig. 5. ... The direction of time can be inferred from the NO<sub>2</sub> trends, as NO<sub>2</sub> concentrations have generally decreased each year over the decade.”**

*Regarding the use of medians for Figures 7 and B2, are there large number of cases with large deviations or extremes?*

We have address this with new text:

**“Fig. 5. ... There is no significant difference between medians and means.”**

**“Fig. 7. ... Medians and means give the same result.”**

**“Fig. B2. ... There is no significant difference between medians and means.”**

*Figure 7 and B2. Is the start of record 2000–2003 or 2001–2004?*

Thank you for your attention. We have corrected caption B2 to read 2001–2004, which are the years plotted.

*Figure B2 caption. The color description is difficult to understand. Is the gray the lighter tint of the yellow lines?*

We have updated the figure to improve the clarity.