

Additional text and equations in response to reviewer 1

In order to compare the calculated instantaneous production rates to the observed $\sum(C_1-C_5 \text{ nitrates})/\sum\text{ANs}$ ratio and $\sum\text{ANs}$ vs. O_x correlation we must use a kind of stepwise integration. The instantaneous and observed slopes or ratios would only be expected to match exactly in the near-field of the source region over which the VOC mixture is invariant. As the plume ages and the VOC mixture changes, the observations reflect a linear combination of all of the previous instantaneous production rather than the single instantaneous slope calculated for a given point. For instance, in the case of the slope of the $\sum\text{ANs}$ vs. O_x correlation, the observed slope after some number, n , of elapsed time intervals (Δt) is given by:

$$M_n = \frac{\sum_{i=1}^n \Delta(O_x)_i * \Delta t_i}{\sum_{i=1}^n \Delta(\sum\text{ANs})_i * \Delta t_i}$$

Clearly for the first time interval, given correct chemistry, the overall slope (M_1) should be similar to the instantaneous slope ($\Delta(O_x)_1/\Delta(\sum\text{ANs})_1$). If the calculated and the observed slopes matched reasonably well for the first time interval we could simply calculate M_2 using the equation above and compare it to the observed M_2 . Since, however, the observed and calculated slopes are entirely dissimilar at point 1, the observed and calculated slopes at point 2 would be different regardless of whether the calculated instantaneous production was correct over Δt_2 because the second step in the calculation is impacted by the error in the first. What we really want to assess is whether the calculated instantaneous production rates during Δt_2 can explain the change between $M_{1\text{obs}}$ and $M_{2\text{obs}}$. To do this we calculate M_n for $n>1$ using the observed M_{n-1} slope as follows:

$$M_{n_calc} \approx \frac{\left(M_{n-1_obs} * \sum_{i=1}^{n-1} \Delta(\sum\text{ANs})_i + \Delta(O_x)_n \right)}{\left(\sum_{i=1}^n \Delta(\sum\text{ANs})_i \right)}$$