

Oxidation flow reactors are widely used in the study of formation and aging of organic aerosols, especially simulating OH radical dominated daytime oxidation process. In this study, the authors extended the use of OFR for studying NO<sub>3</sub> radical initiated nighttime oxidation and the aging process by online producing N<sub>2</sub>O<sub>5</sub> as the source of NO<sub>3</sub>. By model simulation and measurements, the authors investigated the controlling factors of NO<sub>3</sub> exposure, NO<sub>3</sub>:O<sub>3</sub>, NO<sub>2</sub>:NO<sub>3</sub> ratios and provided guidelines for experimental design. I believe it will help researchers in using OFR for nighttime chemistry studies. I recommend to accept it after minor revision.

#### General comments

Regarding the NO<sub>3</sub> estimation equation for the ORF-iN<sub>2</sub>O<sub>5</sub>, I wonder how would multiple generation oxidations influence the estimation of NO<sub>3</sub> exposure. For example, NO<sub>3</sub> radical oxidation of typical BVOCs (isoprene, monoterpenes, sesquiterpenes) produces carbonyls and even products with carbon double bonds. These products are highly reactive toward NO<sub>3</sub> radicals which may affect the NO<sub>3</sub> exposure estimation. However, these are not considered in the KimSim simulations. Secondly, NO<sub>3</sub> oxidation of BVOCs has high SOA yields. I wonder how the uptake of NO<sub>3</sub> and N<sub>2</sub>O<sub>5</sub> by the produced particles affect the simulations.

#### Specific comments:

(1) BBCES measuring the NO<sub>3</sub>. 1) The author stated that “ $I(\lambda)$  and  $I_0(\lambda)$  were the measured transmitted intensities in the presence and absence of NO<sub>3</sub>”. How was the “absence of NO<sub>3</sub>” achieved? 2) The equation regarding the calculation of  $\alpha(\lambda)$  is not correct. The  $\alpha(\lambda)$  in the cavity also contributed by the bath gas beyond the NO<sub>3</sub> radicals. 3) The NO<sub>3</sub> radicals are highly reactive and can easily lose to the walls. What is the transmission efficiency of NO<sub>3</sub> from the OFR to the cavity? 4) Due to different loss rates of NO<sub>3</sub> and N<sub>2</sub>O<sub>5</sub> to the wall, the equilibrium of NO<sub>3</sub> and N<sub>2</sub>O<sub>5</sub> may change. How good is the measured NO<sub>3</sub> concentration in the CRD represent the NO<sub>3</sub> radical concentration in the reactor?

2) The description of the results in section 3.4 is not consistent with the results in the figure.

“First, at  $[\text{O}_3]_{0,\text{LFR}} < 1000$  ppm and  $[\text{NO}_2]_{0,\text{LFR}}:[\text{O}_3]_{0,\text{LFR}} = 0.1$  to 1.8, maximum  $\text{NO}_{3\text{exp}}$  increased with decreasing  $[\text{NO}_2]_{0,\text{LFR}}:[\text{O}_3]_{0,\text{LFR}}$  (Fig. 7a).” It is very hard to see the results in the figure when  $[\text{O}_3]_{0,\text{LFR}} < 100$  ppm. For me, it looks like maximum  $\text{NO}_{3\text{exp}}$  first increase with increasing  $[\text{NO}_2]_{0,\text{LFR}}:[\text{O}_3]_{0,\text{LFR}}$  ratio and then decrease with it, especially when  $[\text{O}_3]_{0,\text{LFR}}$  was in the range of 100-1000 ppm

“Above  $[\text{O}_3]_{0,\text{LFR}} \approx 2000$  ppm,  $\text{NO}_{3\text{exp}}$  was less sensitive to  $[\text{NO}_2]_{0,\text{LFR}}:[\text{O}_3]_{0,\text{LFR}}$ .” This is true except for  $[\text{NO}_2]_{0,\text{LFR}}:[\text{O}_3]_{0,\text{LFR}}=2.0$

“Second, maximum  $\text{NO}_3:\text{O}_3$  increased with increasing  $[\text{NO}_2]_{0,\text{LFR}}:[\text{O}_3]_{0,\text{LFR}}$  (Figure 7c).” This statement is true only when the  $\text{O}_3$  was above 1000 ppm, even get rid of the results from  $[\text{NO}_2]_{0,\text{LFR}}:[\text{O}_3]_{0,\text{LFR}}=2.0$ .

“conversion of  $\text{O}_3$  to  $\text{N}_2\text{O}_5$  inside the LFR” I fell more comfortable to say “conversion of  $\text{O}_3$  to  $\text{O}_2$  inside the LFR”.

3) The authors tried to investigate the  $\text{RO}_2$  fate and considered “ $\text{RO}_2$  react with  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{NO}_3$ ,  $\text{HO}_2$ , or other  $\text{RO}_2$  to generate alkoxy (RO) radicals, peroxy nitrates ( $\text{RO}_2\text{NO}_2$ ), hydroperoxides or organic peroxides, and may additionally undergo autooxidation via sequential isomerization and  $\text{O}_2$  addition.” Recent studies by Berndt et al. (2018) revealed that self- and cross-reaction of  $\text{RO}_2$  radicals would produce dimers effectively. How could this process affect the fate of the  $\text{RO}_2$  radical?

1. Berndt, T.; Mender, B.; Scholz, W.; Fischer, L.; Herrmann, H.; Kulmala, M.; Hansel, A., Accretion Product Formation from Ozonolysis and OH Radical Reaction of alpha-Pinene: Mechanistic Insight and the Influence of Isoprene and Ethylene. Environ. Sci. Technol. 2018, 52, (19), 11069-11077.

2. Berndt, T.; Scholz, W.; Mentler, B.; Fischer, L.; Herrmann, H.; Kulmala, M.; Hansel, A., Accretion Product Formation from Self- and Cross-Reactions of  $\text{RO}_2$  Radicals in the Atmosphere. Angew Chem Int Edit 2018, 57, <sup>1</sup>, 3820-3824.