

**Comments by Reviewer #4:**

(Page/Line numbers refer to the previous version of the paper; they are simply copied from the reviewer report)

Page 11-12:

I think the reviewer may have referred to studies by Parrish et al., such as Parrish, J. Geophys. Res. 2002 (10.1029/2001JD000720), or Atmos. Environ. 40 (2006) 2288–2300

**Author's reply:**

**The reference of Parrish et al. (2002) was added in line 276. Thanks for the information.**

Page 11, line 4-5 and following:

I think your response was great and includes important information; however, I did not find if/where you added it to the revised manuscript.

**Author's reply:**

**Apologies for the insufficient explanation. It is feasible to explain that the majority of organics was made of carboxylic acid that was fully or partially neutralized by ammonium because 1) there was more than enough  $\text{NH}_4^+$  to neutralize sulfate and nitrate (stated in line 295-299), 2) the  $r^2$  between  $m/z$  44 and  $\text{NH}_4^+$  was higher than the  $r^2$  between  $m/z$  44 (or organics) and sulfate (Table 2), and 3) the PMF analysis showed that LV-OOA, which can be estimated according to the marker of  $m/z$  44, is the major component of organics (line 341-342). We revised the discussion (line 289-305) because your 2<sup>nd</sup> comment made us realize that our explanation for the neutralization by ammonium was confusing. Thanks for your comment again.**

Figure 9:

“...If you meant the OM/OC ratios, those ratios for HOA are zero (Table 1)”

I got confused about your response here. 1) I don't see any OM/OC ratios in Table 1, 2) How can an OM/OC ratio be 0 (i.e.  $\text{OM} = 0$ )? Do you mean for very low oxidized material  $\text{OM/OC} \sim 1$ ?

**Author's reply:**

**Figure 9 you refer would be Figure 8 in the revised manuscript. Apologies for**

referring the incorrect table and explaining the OM/OC ratios incorrectly. The table was supposed to mean Table 3. The OM/OC should not be zero. Zero we meant was f44 for HOA component.

Back to your original critics, we double checked the OM/OC ratios for HOA and LV-OOA components. We then found that the calculation for OM/OC was incorrect. Correct OM/OC ratios for HOA and LV-OOA are 1.7 and 4.2, respectively. The ratios in Table 3 and the text (line 353) were revised accordingly. This change also required a change in the parameter “a” in the model calculation. To have the same fit as the previous plot, the “a” value needed to change to 0.025. This change was made in Table 3 and line 559. Figure 8 was also updated accordingly.

## Editor comments

Here the line numbers refer to your revised manuscript.

### Comments:

l. 220: Can you give numbers for the LDL values? Has LDL been defined before?

**Author's reply: LDL for NO<sub>x</sub> and NO<sub>y</sub> means < 0.006 ppbv. "LDL, <0.006" was added in line 215.**

l. 265/6: I do not understand the argument 'Because the processes are physical we expect that the order of NO<sub>y</sub> lifetime in our study is similar.'

**Author's reply:**

**The explanation was revised to give our point of view more specifically (line 266-271).**

l. 330: Can you add a reference to the statement that humic-like substances have high OM/OC ratios?

**Author's reply: A statement "Based on the AMS reference mass spectra available from the web site previously referred, ..." was added in line 357-358.**

l. 383: Not clear. R4 as it is written does not include any pH dependence. Why does the fact that sulfate was neutralized by ammonia makes R4 negligible?

**Author's reply: Apologies for the poor explanation. The information is now given more specifically (line 406-410).**

l. 572: Add the value and reference for the rate from the New England Air Quality Study here

**Author's reply:**

**The rate,  $1.3 \times 10^{-9} \times [\text{OH}] \text{ h}^{-1}$ , was added (line 591).**

l. 530: Not clear what you mean

"As two members, HOA and LV-OOA, had similar f<sub>44</sub> values, the indicator **did** not work" or

"If two members, HOA and LV-OOA, had similar f<sub>44</sub> values, the indicator **would** not work"

**Author's reply:**

**Sorry for confusing. "HOA and LV-OOA" was removed because the "two members" are not necessarily HOA and LV-OOA. As you suggest, we revised the**

**sentence to as follows: “If two members had similar f44 values, the indicator would not work.” (line 548-549).**

#### **Technical comments**

l. 93: sometimes

**Author’s reply:**

**Corrected (line 93).**

l. 128; 130 min: under 1 min averaging time → with 1 min averaging time

**Author’s reply:**

**Corrected (line 128 and 130).**

l. 144: 2,5 → 2.5

**Author’s reply:**

**Corrected (line 144).**

l. 152: ‘standard deviation’ can be omitted as SD has been defined before

**Author’s reply:**

**The term was removed (line 152).**

l. 258: originate → originates

**Author’s reply:**

**Corrected (line 264).**

l. 268: in the similar order to → on the similar order of

**Author’s reply:**

**Corrected (line 272).**

l. 279: “Nevertheless of such... “ – do you mean “Despite of...”?

**Author’s reply:**

**It was changed to “Despite” (line 284), to be consistent with usage of “Despite” in other places in the text.**

l. 282: sinking → sink

**Author’s reply:**

**“sinking process” was changed to sink (line 287).**

l. 327: at some extent → to some extent

**Author's reply:**

**The sentence including “at some extent” was removed from line 355 because incorrect information, which was pointed out by the Reviewer #4, was presented.**

l. 382: remind → remember

**Author's reply:**

**“one should remind” was removed from the sentence (line 408).**

l. 393: 4 times as fast as → four times faster than

**Author's reply:**

**The sentence was removed due to the removal of the lifetime comparison (see the 5th in “Additional revision” below).**

l. 424: Robert → Roberts

**Author's reply:**

**Corrected (line 443).**

l. 437: under → at

**Author's reply:**

**Corrected (line 456).**

l. 507: another → other

**Author's reply:**

**Corrected (line 526).**

l. 524: ‘The greater extent of reaction processing proceeds..’ – please reword

**Author's reply:**

**“processing” was removed (line 543).**

l. 573: implying → imply

**Author's reply:**

**Corrected (line 592).**

l. 575: increasing → increase

**Author's reply:**

**Corrected (line 594).**

Figure 7, l. 860: as function  $\rightarrow$  as a function

**Author's reply:**

**The figure captions for Figure 7 and 8 were corrected accordingly.**

#### **Additional revision**

- 1. The tile was not revised in the last revised manuscript. The title was updated.**
- 2. The averaging time and DL value provided for CO was found to be incorrect. These values are corrected in line 130-131 in the most updated manuscript.**
- 3. We may have misunderstood the original comment from the reviewer #4, "Figure 9. Why is the f<sub>44</sub> of HOA exactly zero? OA/OC for this PMF component has an O to C ratio approximately equal to one." The O to C ratio of one seems to be the 43/44 ratio shown in Figure S-14. We looked into the results of PMF analysis and found a contribution of semi-volatile oxygenated organic aerosol (SV-OOA) sometimes. The 43/44 ratio of one can be explained by a combination of HOA (or SV-OOA) and LV-OOA. This discussion was revised in line 331-338.**
- 4. An additional figure supporting the additional statement for PMF analysis was added to the supporting information (Figure S-15).**
- 5. Comparison of lifetimes calculated for the R1 and R2 channels were removed (line 384-394 in the previous revision) because the discussion was found to be redundant and meaningless after stating that the series of R2-R4 channels was negligible based on the clue by neutralization of sulfate and nitrate by ammonium. The discussion for the negligible nocturnal sink of NO<sub>x</sub> was revised (line 406-422).**
- 6. Figure 2 and 3 were replaced with more legible ones.**