

Interactive comment on “A case study of aerosol processing and evolution in summer in New York City” by Y. L. Sun et al.

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Response to the comments of referees

We thank the reviewers for their thoughtful comments. The manuscript has been revised accordingly. Listed below are our point-by-point responses to each reviewer's comments, which are repeated in italic.

Response to Referee #1

Sun et al. reported an observation on an aerosol processing and evolution event from 21-22 July 2009 in New York City. A HR-AMS collocated with many other instruments

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were employed to obtain the data of aerosol, trace gases, VOCs, etc synchronously. An aerosol evolution process with three clear stages of (1) aerosol wet scavenging, (2) nighttime nitrate formation, and (3) photochemical production and evolution of secondary aerosol processes were recorded. Elaborate data analyses and discussion are presented, interpreting the evolution process well.

Aerosol wet scavenging in an open system is a complicated and dynamic process. The O/C of OA may have some certain correlations with the wet scavenging rate generally. However, in this special case, authors should explain well why HOA, COA, nitrate, and chloride showed little changes during the aerosol wet scavenging and present more discussion about the positive scavenging rates of SV-OOA, COA, and HOA.

[Response]: Following the reviewer's suggestions, we have expanded the discussions on the reasons for the little changes of species during the aerosol scavenging period and the positive scavenging rates of SV-OOA, COA, and HOA. This includes:

“The small variation of HOA is also likely due to the constant inputs from local emissions, in agreement with the variations of primary gaseous tracers for traffic emissions, e.g., NO, NO₂, and CO₂.”

“Nitrate, despite its high hygroscopicity, showed almost no removal (-0.1%). The apparent low scavenging rate is likely due to the competing effects between rainfall scavenging and the continuous, but relatively low, input of nitrate during this stage. Indeed, the photochemical production rate of HNO₃ (Sect. 3.2) showed a slight increase after 10:00, indicating the formation of HNO₃ that can react with NH₃ to form NH₄NO₃. Another possible reason for the persistently low concentration of nitrate is that not all fine aerosols were efficiently scavenged by rain drops (Andronache, 2003). This might also explain the low yet non-zero concentration of sulfate observed at the end of this stage even though the scavenging rate of sulfate is high.”

“The primary OA factors (COA and HOA), as well as EC, showed slight increases despite rain scavenging, part of which was due to local emissions. Especially, the increase of COA was associated with enhanced local cooking activities in the evening.”

"OA on average became less oxidized due to the much faster scavenging of OOA than HOA." (Page 25761, line12-13) may not be a comprehensive conclusion if there was continuous input of HOA from local emissions. Nevertheless, this paper is suitable for publication in Atmospheric Chemistry and Physics after the following comments have been taken into account.

[Response]: We agree that the increase of O/C for bulk OA was due to both faster scavenging of OOA and continuous input of POA from location emissions. The text has been revised to reflect this point.

Specific comments

Page 25752, line 17: "semi-volatile OOA" should be "SV-OOA" for it is the second time used in the paper.

[Response]: "semi-volatile OOA" was replaced by "SV-OOA"

Page 25752, line 24: The usage of the phrase "the sometimes lack" looks not proper.

[Response]: The sentence was revised as "which might explain, sometimes, the lack of correlations between LV-OOA and sulfate"

Page 25753, line 18: The sentence "As the..., there is..." seems not logical, Suggest using "Though" instead of "As".

[Response]: "As" was changed to "Although"

Page 25754, line 4: "revisited" is not a fine word to describe the action.

[Response] “revisited” was replaced by “returned to”. The sentence now reads “In summer 2009, we returned to Queens College (QC) in New York City (NYC), previously an EPA supersite during PMTACS-NY, with an Aerodyne High-Resolution Time-of-Flight AMS (HR-AMS; DeCarlo et al., 2006) to study submicron aerosol chemistry and processes (Sun et al., 2011a).

Page 25756, line 6: It will be better to offer the full names of “OC” and “EC” as they appear at the first time in the text.

[Response]: “OC” and “EC” were spelled out.

Page 25757, line 12: “Figure S1” should be “Fig. S1” for consistent with others.

[Response]: We keep “Figure S1” in the revised manuscript following the requirements of ACP, i.e., using “Figure” instead of “Fig.” when a sentence starts with “Figure xx...”.

Page 25762, line 20 “BC” should be “EC” according to Fig. 1.

Page 25766, line 11: “~ 0.7” should be “– 0.7”.

Page 25766, line 13: “~ 0.5” should be “– 0.5”.

Page 25766, line 26: “ $2\text{NH}_4\text{NO}_3(\text{NH}_4)_2\text{SO}_4$ ” should be “ $2\text{NH}_4\text{NO}_3 \bullet (\text{NH}_4)_2\text{SO}_4$ ”.

Page 25767, line 20: “BC” should be “EC”.

Page 25768, line 14: “secondary OA” should be “SOA”.

Page 25769, line 2: “that that” should be “to that”.

[Response]: Corrected

Page 25778, Fig. 1: The labels of the plots (a, b, c...) are upside-down; The line for “Chl” in Fig. 1 (b) is hard to see; The caption of Fig. 1 is disordered and confusing, It will be better to rewrite the caption according to the sequence of a, b, c. ...

[Response]: Following the reviewer’s suggestions, we revised the labels of Fig. 1 and

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the caption as well. Also the time series of Chl was enhanced by a factor of 5 in the figure for clarity.

Page 25783, the caption of Fig. 6: Add "and" before "OA/ Δ CO₂".

[Response]: "and" was added before "OA/ Δ CO₂"

Response to Referee #2

This paper reports an aerosol processing event in summer in New York City. AMS offers highly time resolved information on the evolution of major aerosol components during three major stages wet scavenging, nitrate formation and photochemical production of SOA. This is a well-written paper and could be accepted by ACP after the following issues are addressed

1) For the first stage, the authors explained well about the wet scavenging of ammonium sulfate mostly based on its high hygroscopicity. However, nitrate (What is the major form of nitrate during this stage, ammonium nitrate?), which is a well-known hygroscopic material, showed little change in concentration during this step. Why? The scavenging rate of nitrate should be presented in Table 1 with more discussion.

[Response]: The ion balance analysis indicates that particulate nitrate was fully neutralized by ammonium during this stage. According to the E-AIM model results shown in Fig. 4, nitrate existed in the aqueous-phase completely during the first stage.

Nitrate remained at low concentration ($\sim 0.2 - -0.7 \mu\text{g m}^{-3}$) and showed little variation during this stage despite effective wet scavenging of hygroscopic species. One of the main reasons is likely the continuous, albeit relatively low, input of nitrate formed from the reaction of HNO₃ with NH₃ under the conditions of low temperature and high

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relative humidity. Indeed, as shown in Fig. 4, $\text{NO}_2 \times \text{UV}$, a surrogate of photochemical production rate of HNO_3 , showed a slight increase after 1000, indicating the formation of HNO_3 that can be further neutralized by NH_3 to form NH_4NO_3 . In addition, depending on rainfall rates and rain drop size distributions, a fraction of the submicron particles may survive the scavenging (Andronache, 2003), explaining the observation of low yet non-zero concentrations of nitrate, sulfate, and OOA during rainfall events. Following the reviewer's suggestions, we have included the scavenging rate of nitrate in Table 1 and expanded the discussions on the scavenging of nitrate in Section 3.1 (Paragraph 2) in the revised manuscript.

2) For the second stage, the heterogeneous reaction followed by neutralization was proved to be the dominant formation mechanism of nitrate. This night-time nitrate formation had been reported many times in previous studies. The authors should step forward a little and discuss the possible reasons for the high concentrations of NO_2 and NO and low concentration of O_3 during this period.

[Response]: The high concentration of NO_2 and NO during the second stage is likely due to local traffic emissions, which is further supported by their similar trends to other tracers for traffic emissions such as EC and HOA. The low level of O_3 is likely due to the reactions with NO and NO_2 ($\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$; $\text{NO}_2 + \text{O}_3 \rightarrow \text{NO}_3 + \text{O}_2$). We discussed the possible reasons for high NO_x and low O_3 in Section 3.2.

3) More explanation should be given for Fig. 5 both in caption and text. The definition of dash lines may be well known in AMS society but not for general audience.

[Response]: In response to the reviewer's comment, we expanded the descriptions of dash lines in Fig. 5. Now it reads "Fig. 5. (a) Relationship between f44 (fraction of m/z 44 in the total organic signal) and f43 (fraction of m/z 43 in the total organic signal) during 21 – 22 July. The f44 vs. f43 relationships for five OA components identified during this study are also shown. The dash lines in (a) refer to a triangular

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region that encompasses ambient OOA factors determined from PMF analyses of 43 AMS datasets (Ng et al., 2010). More details are discussed in the main text. (b) Van Krevelen diagram for OA from 21-22 July and the five OA components identified from PMF analyses. The dash lines in (b) indicate the changes of H/C against O/C due to adding specific functional groups to an aliphatic carbon (Heald et al., 2010).”

References:

Andronache, C. Estimated variability of below-cloud aerosol removal by rainfall for observed aerosol size distributions, *Atmos. Chem. Phys.*, 3, 131-143, 10.5194/acp-3-131-2003, 2003.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 25751, 2011.

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