

Response to reviewer 2

We thank the reviewer for careful comments and suggestions. Following is our response to the comments:

Comments RC2:

In the manuscript the authors apportioned the primary and secondary sources of the organic aerosols using a chemical mass balance (CMB) and trace yield methods based on 144 kinds of quantified organic species, including both primary and secondary tracers. The effectiveness of control measured on primary and secondary sources were assessed based on the obtained results. Back trajectory cluster analysis was also conducted to evaluate the influences of air mass directions on the organic aerosol sources. Environmental factors, such as temperature, O₃ concentration, aerosol liquid water content, and particle acidity were also investigated to elucidate the formation mechanisms of secondary organic aerosols. The topic of the manuscript fits very well into the Atmospheric Chemistry and Physics and the manuscript is well written. Generally, I recommend the publication of the manuscript.

However, there are some technical details that might change the conclusion of the manuscript, which I think need to be addressed before its publication.

Main Comments

*1. The authors spend the whole section 4.3 “Influencing factors for secondary organic formation in the summer of Beijing”, discussing the factors that could influence the anthropogenic SOC (Figure 4). To get their point, they did correlation plot of the anthropogenic SOC loading with different factors and positive slope indicating enhancing effects. I found this not reasonable. What the authors really need is “multivariate analysis” or “multivariate regression”. Otherwise, one factor could have influenced the behavior of the other factor and change the sign of the slope, leading to an opposite conclusion. For example $y = f(x_1, x_2) = x_1 - 0.5 * x_2$. y is positively correlated with x_1 , but negatively correlated with x_2 . You made some measurements at $x_1 = 1, x_2 = 0$ and $x_1 = 2, x_2 = 1$. The two y 's you will obtain are 1 and 1.5. Then based on the authors method, one will obtain y is positively correlated with x_1 (with slope of 0.5) and x_2 (with slope of 0.5).*

We thank the reviewer for the comments.

What we want to do in section 4.3 is to roughly discuss the influencing factors that can have an impact on the SOC concentration, thus shed light on further study to concentrate on the influencing factors concerning the SOA formation. So we use univariate analysis to see which factor may influence the apportioned SOC and see the correlation

between the potential influencing factors and the apportioned SOC. The correlation between different parameters could at least enlighten us of the influencing factors for SOA formation in megacities such as Beijing under the complex air pollution conditions.

Besides, we did the multivariate analysis as the reviewer suggested. The multiple regression analysis was used to investigate the relationship between SOC and water content, H^+ , temperature, and ozone concentration. The multiple regression equation was as following:

$$SOC = 0.5495 + 0.052 \times \text{water content} + 5.24 \times H^+ + 0.01085 \times \text{temp} + 0.01054 \times O_3$$

The correlation coefficient $R=0.73$. All the influencing factors have positive impact on the SOC concentration. According to our results, H^+ concentration has significantly great impact on SOC formation.

Anyway, all these influencing factors can interact with each other. Therefore, the multivariate analysis also has large uncertainties.

2. The authors did show in Figure 1 that the governmental control changes the Organic aerosol apportionment a bit, however, the total organic aerosol loading does not change much, or even increased (from 8.9 ug/m³ to 11.0 ug/m³) (as shown in Table 1 too). The total PM_{2.5} loading has decreased from 92.3 ug/m³ to 45.5ug/m³. Then this leaves the reader wonders what have been decreased mostly? The sulfate?

Nitrate? Ammonia? Or something else. The authors need to add the loading of these into Table 1. The decrease of EC from 3.3 $\mu\text{g}/\text{m}^3$ to 1.8 $\mu\text{g}/\text{m}^3$ is not enough to explain the more than 40 $\mu\text{g}/\text{m}^3$ decrease in $\text{PM}_{2.5}$.

We agree with the reviewer. Additional discussion about other compounds, i.e. inorganic components, was included in the revised text.

We could see from table 1 that after the government took control strategies, the concentrations of $\text{PM}_{2.5}$, EC decreased significantly since 2008. However, the OC concentrations didn't show the same tendency with $\text{PM}_{2.5}$ and EC. To elucidate the reasons for the dramatic decrease of $\text{PM}_{2.5}$, we compared the data of the main inorganic water soluble ions i.e. sulfate, nitrate and ammonia (relevant data has been added to table 1). Results showed that the averaged concentration of water inorganic water soluble ions decreased from 2008, with sulfate decreased from 35.6 $\mu\text{g}/\text{m}^3$ to 4.7 $\mu\text{g}/\text{m}^3$, nitrate decreased from 7.9 $\mu\text{g}/\text{m}^3$ to 2.4 $\mu\text{g}/\text{m}^3$, ammonia decreased from 15.2 $\mu\text{g}/\text{m}^3$ to 3.5 $\mu\text{g}/\text{m}^3$. The significant decrease of SNA and EC confirmed the effectiveness of the drastic measures taken by the government. Therefore, the reduction of fine particulate matter was mainly due to the well controlling of the EC and inorganic particulate matter such as sulfate, nitrate and ammonia, especially the dramatic decrease of sulfate (86.8% from 2008 to 2016).

The relevant data of SNA has been added to table 1, and the discussion

about the decrease of $PM_{2.5}$ was as follows (line 222-228) “Relevant data of main WSICs (sulfate, nitrate and ammonia) during 2008 to 2016 were also included in table 1 to better elucidate the drastic decrease of fine particulate matter in recent years. Results showed that the daily average concentration of WSICs decreased from 2008 to 2016, with sulfate decreased from $35.6 \mu\text{g}/\text{m}^3$ to $4.7 \mu\text{g}/\text{m}^3$, nitrate decreased from $7.9 \mu\text{g}/\text{m}^3$ to $2.4 \mu\text{g}/\text{m}^3$, ammonia decreased from $15.2 \mu\text{g}/\text{m}^3$ to $3.5 \mu\text{g}/\text{m}^3$. The significant decrease of WSICs confirmed the effectiveness of the control measures taken by the government” and “Therefore, we could draw a conclusion that the drastic decrease of fine particulate matter in Beijing was mainly due to the well-controlled EC and WSICs, with negligible contribution of OC”.

Besides the above two comments, I also have some minor comments as listed below.

1. Line 119, by “filters” does the authors mean “quartz filter” only. Or the authors analyzed both “quartz filter” and “Teflon filter”.

Thank you for your comment.

The “filters” mentioned here was referred to quartz filters only. As is mentioned above, the four-channel samplers (TH-16A, Tianhong, China) consisted of three quartz filter channel and one Teflon filter channel.

Teflon filter was weighed and used to calculate the concentration of PM_{2.5} and analyze the water-soluble inorganic compounds. The quartz filters were used to analyze the EC, OC and the particulate organic matters. Here, the “filters” referred politically to quartz filters.

The manuscript has been altered from “The filters were then ultrasonically extracted with methanol: dichloromethane (v:v=1:3) solvent in water bath (temperature < 30 °C) for 3 times” to “The quartz filters were then ultrasonically extracted with methanol: dichloromethane (v:v=1:3) solvent in water bath (temperature < 30 °C) for 3 times” to avoid ambiguity (line 110).

2. In Figure S7, are the vertical lines the measurement error bars or they indicate the daily ranges? As this could change the statement of line 270 stating that hope at PKUERS site were much higher than that of CP.

We thank the reviewer for the comment.

The vertical lines represent the standard deviation of the daily concentrations. For comparison, we compared the daily average values for simplification and thus stated that the hopanes at the urban site PKUERS were higher than that of CP.

The relevant context “For all the species, the histogram showed the average daily concentrations with error bars representing the standard

deviations” has been added to the manuscript (line 256-257)

3. *Line 305, the concentrations of what in CP were lower than that of PKUERS?*

Thank you for your comment.

It's the concentration of 2,3-dihydroxy-4-oxopentanoic acid that was lower in CP compared with PKUERS. We revise this sentence to make it clear: “However, the 2,3-dihydroxy-4-oxopentanoic acid concentrations in CP were lower than that of PKUERS...” (line 318-319)