

## Dear Editors and Reviewers:

Thank you for your letter and for the reviewers' comments concerning our manuscript entitled "Non-polar organic compounds in aerosols in a typical city of Eastern China: Size distribution, gas-particle partitioning and tracer for PM<sub>2.5</sub> source apportionment" (Ref: acp-2017-908). These comments are valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have studied comments carefully and have made correction, the correction in the manuscript was marked-up with GREEN colour which we hope meet with approval. The main corrections in the paper and the responds to the reviewer's comments are as flowing:

Responds to the editors' and reviewers' comments:

## Anonymous Referee #3

**Received and published: 28 March 2018**

**The manuscript has the potential to add to the available body of evidence. This work details the size distribution, gas-particle partitioning and source apportionment of airborne PM<sub>2.5</sub>-associated non-polar organic compounds in one city of eastern China. In general, I recommend that the manuscript be accepted pending some major revisions as outlined below.**

### **1. How about the air quality of Jiujiang City? Is there any public data about the air quality there?**

**Response:** There was a small number of published researches about the atmosphere quality of Jiujiang City. According to Yang's (2016) research, PM<sub>2.5</sub> was the dominant atmospheric pollutant in Jiujiang City, which ranged from 31 to 196  $\mu\text{g m}^{-3}$  in 2014-2015, and the highest abundances of aerosols was usually occurred during October to January of the next year.

According to a recent research program "Source Apportionment of PM<sub>2.5</sub> and VOCs, and Control and Management of Jiujiang City" (data not published), the average concentrations of PM<sub>2.5</sub> and O<sub>3</sub> in Jiujiang City were 45, 51, 50  $\mu\text{g m}^{-3}$  and 43, 55 and 61  $\mu\text{g m}^{-3}$  in 2014–2016, respectively. PM<sub>2.5</sub> and O<sub>3</sub> both showed a gradual rising trend, with annual growth rate of 4.3% and 20.9%, respectively. As for chemical constituents, the secondary inorganic aerosols occupied 53.4% of the PM<sub>2.5</sub> concentration, while OC occupied of 22.1%. Factors extracted by PMF, CMB receptor models showed that coal combustion, vehicle exhaust, industrial production (petrochemical) and dust contributed 31.4%, 14.3%, 13.1% and 11.2%, respectively. The  $\Sigma_{103}\text{VOCs}$  (volatile organic compounds) concentration was 11.5-197.8 ppbv, with average value of 78.20±49.92 ppbv, which is close to in megecities of Beijing and Shanghai in China. Aromatic hydrocarbon was the primary VOCs component, which occupied of 34.6% of the total VOCs. The source identification

of VOCs extracted by PMF showed the largest contributor was the solvent/coating emission (21.1%), followed by incomplete combustion which accounting for 19.8%.

**2. Please cite the references at the end of the sentence, not in the middle.**

*Response:* According to referee's suggestion, nearly all these references cited in the middle were revised as be cited at the end of the corresponding sentence in the revised manuscript. For example, line 30-31 "In recent years, severe atmospheric pollution characterized by haze has been occurring in developing countries (Yadav et al., 2013;Wang et al., 2015), affecting visibility, optical radiation and human health (Shen et al., 2015;Sulong et al., 2017)." was changed to "In recent years, severe atmospheric pollution characterized by haze has been occurring in developing countries, affecting visibility, optical radiation and human health (Yadav et al., 2013;Shen et al., 2015;Wang et al., 2015;Sulong et al., 2017)." in lines 30-31 in the revised manuscript.

However, several references were kept cited in the middle of the sentence due to the special structure of sentence. For example, in line 243-244 "This was consistent with the measurement results of NPOCs in Pearl River Delta (PRD) (Wang et al., 2016) and South China Sea (Zhao et al., 2016) in China, with percentages of 0.1–4.2% and 0.8–1.7%, respectively." was not changed.

**3. Line 97, please give the size of the quartz fiber filter used. And how was the air flow rate of medial-volume air sampler? Detailed information should be given.**

*Response:* According to referee's suggestion, the size of particle filter and sampler flow were added to the corresponding descriptions in line 97 of the original manuscript, changed as "All PM<sub>2.5</sub> filter samples were collected using medial-volume air samplers (YH-5, Qingdao, China), at a flow rate of 100 L min<sup>-1</sup>. Particles were collected on quartz fiber filters (GE Whatman, 1851-090, England, UK) with a diameter of 90 mm, each for a duration of 23 h." in lines 98-100 in the revised manuscript.

**4. Line 98, did you mean that each sample was performed for five continuous days? Did you consider the filter would be oversaturated for such long sampling time?**

*Response:* According to referee's suggestion, the statements the sampling duration 23 h for each single sample was added, and in general the filter would not be over-saturated for such a sampling time in this study. The revised description was "Particles were collected on quartz fiber filters (GE Whatman, 1851-090, England, UK) with a diameter of 90 mm, each for a duration of 23 h." in lines 99-100 in the revised manuscript.

## 5. How many samples did you get totally?

**Response:** According to referee's suggestion, the total sampling number of PM<sub>2.5</sub> was added in the revised manuscript, as "A total of 137 PM<sub>2.5</sub> valid samples were collected, as 18 samples were invalid or missing due to the bad weather or power problems." in lines 103-104 in the revised manuscript.

## 6. Line 107, why did you choose two sizes of quartz fiber filters? Was one for medial- volume air sampler, while other for MOUDI? Modify clearly and suitably.

**Response:** This MOUDI sampler was designed to collect particles with diameters of 0.056-18.0 and 0.010-0.056  $\mu\text{m}$  using 47 and 90 mm filters, respectively. According to referee's suggestion, line107 "Two kinds of quartz fiber filters (diameters of 47 and 90 mm, respectively) were prebaked at 550 °C for 4 h," was changed to "Two kinds of quartz fiber filters of diameters of 47 and 90 mm were used to collect particles with diameter of 0.056-18.0 and 0.010-0.056  $\mu\text{m}$ , respectively. All the filters were prebaked at 550 °C for 4 h." in lines 112-113 of the revised manuscript.

## 7. Line 129, why didn't you choose SIM mode?

**Response:** The TD-GC/MS method used for the aerosol associated NPOCs analysis in this study, was referred to studies conducted by Ho and Yu (Ho and Yu, 2004; Ho et al., 2008), and with several parameter be modified. Numerous previous researches which studied NPOCs via using TD-GC/MS selected SCAN mode instead of SIM mode, see following Table R1.

**Table R1** References related to scan mode for NPOCs by TD-GC/MS approach

Location	Instrument	SCAN or SIM	Target compounds	Reference
British Columbia, Canada	TDU+Agilent GC6890/MS5973	SCAN: 50-1000 amu	PAHs, n-alkanes, biomarker	Ding et al., 2009
Hong Kong, China; Fresno, CA, USA	TDU+Agilent GC6890/MS5973	SCAN: 50-650 amu	PAHs, n-alkanes, iso/anteiso-alkanes, hopanes, steranes, branched alkanes, cyclohexanes, alkenes, pathalates	Ho et al., 2008
Hong Kong, China;	TDU+Agilent GC5890/MS5791	SCAN: 50-650 amu	PAHs, n-alkanes,	Ho and Yu 2004
Six cities in China and Japan	TDU+Agilent GC6890/MS5975	SCAN: 50-650 amu	PAHs, n-alkanes, hopanes, steranes, pathalates	Ho et al., 2011
Delhi, India	TDU+Shimadzu GC/MS QP2010 Plus	SCAN: 40-900 amu	PAHs, n-alkanes	Yadav et al., 2013
Pearl River Delta, China	TDU+Agilent GC/MS	SCAN: 50-650 amu	PAHs, n-alkanes, hopanes, steranes	Wang et al., 2016
Jiujiang, China	TDU + Shimadzu GC/MS QP2010 Plus	SCAN: 50-500 amu	PAHs, n-alkanes, hopanes, steranes	This study

Accordingly, the description of scan mode and determining NPOCs species was revised as “The MS was operated in scan mode with mass range was m/z 50–500, and scanned at 0.5 s/scan (Ho et al., 2008;Yadav et al., 2013). The ion was produced from electronic impact ionization (EI) at 70 eV, and then was separated by high performance quadrupole mass filter. Species identification was achieved via comparing the mass spectra and retention times of the chromatographic peak with the corresponding authentic standards.” in lines 140-143 of the revised manuscript.

**8. Line 143, "PM<sub>2.5</sub> samplers were placed on the rooftop at EM site with distance between any two samplers <3 m, collecting for 12 h, then the added mass was calculated." This sentence is not written in proper English, and it's not clear what point is being made here. I recommend deleting this sentence.**

**Response:** According to referee’s suggestion, the sentence in line 143 was shortened and revised as “PM<sub>2.5</sub> samplers were placed on the building rooftop of Jiujiang Environmental Monitor Station, each two within distance of <3 m.” in line 160-161 in the revised manuscript.

**9. When did you add the internal standards?**

**Response:** The internal standards were added to the sample after sample be added with glass-wool plugs. According to referee’s suggestion, “The internal standards of n-tetracosane d<sub>50</sub> (n-C<sub>24</sub>D<sub>50</sub>), naphthalene-d<sub>8</sub>, acpnaethene-d<sub>10</sub>, phenanthrene-d<sub>10</sub>, and chrysene-d<sub>12</sub> were spiked into each sample, through a pipette with a long thin tip. This was done to account for the loss of components from sample filters associated with the instrument instability due to changes in laboratory environmental conditions. After the evaporating of solvent from internal standard was conducted via air drying for several seconds, the TD tubes were capped and put into a sampler holder.” in lines 128-133 in the revised manuscript

**10. Should give some short information on SRM 2266, SRM 2260A and SRM1494.**

**Response:** According to referee’s suggestion, the description of NPOCs standards were revised as “The NPOCs standards used were National Institute of Standards and Technology (NIST, USA) Standard Reference Materials (SRM), including SRM 2260A, SRM 1494 and SRM 2266 for 35 PAHs, 30 n-alkanes and 10 hopanes/steranes, respectively.” in lines 165-167 in the revised manuscript.

**11. I can't find you any statement about the recovery. Did you subtract the average blank level from all samples? Please clarify.**

**Response:** According to referee’s suggestion, the statement of recovery was added in this revised manuscript, depicted

as “Recovery experiment were conducted to improve the desorption of targeted compounds from filters and experimental detection. The analytical recovery was calculated via spiking a known amount of the solution to blank filter, and most compounds were recovered with recovery efficiency >90% except for several light molecule weight species.” in lines 177-1780 in the revised manuscript.

The blank experiments conducted in this study were composed of filed blank, transport blank and laboratory blank experiments, and the final reported data were subtracted the average blank results, just as depicted “Field blanks were collected by keeping blank filters in the sampler for the same duration at sampling site. Additionally, both transport and laboratory blank filters were analyzed, and all the data reported in this study were corrected according to the results.” in line 161-164 in the revised manuscript.

**12. Please state what the levels were in the processing blanks, and how thy compared to your lowest standard.**

**How do you define the LOQ?**

*Response:* Processing blank is processed through some or all equipment used for collecting and processing environmental samples. We have conducted processing blank experiments in this study, results showed that the contamination in the processing period was negligible, which may be due to the experiment were conducted in the clean laboratory room. The lowest standard values were compared with these blank values, which met related requirement. The limit of quantification (LOQ) was calculated as ten times values of of S/N (signal/noise).

**13. Did you include spike blanks? How was accuracy of the method evaluated?**

*Response:* We have not conducted spike blank experiments in this study, due to their similar functions to processing blanks. Despite spike blanks were not included, filed blanks, laboratory blanks and processing blanks all were conducted and received rather good results, which suggesting our experiment was receivable.

The description of the accuracy of the method was added in the revised manuscript, depicted as “The accuracy of the method was evaluated by reproducibility of the standard and selected samples ascertained by processing in quintuplicate, and results suggest the analytical precision was better than 5%.” in line 180-181 in the revised manuscript.

**14. Line 215, is it your own data or cited data from reference? How did you determine OM?**

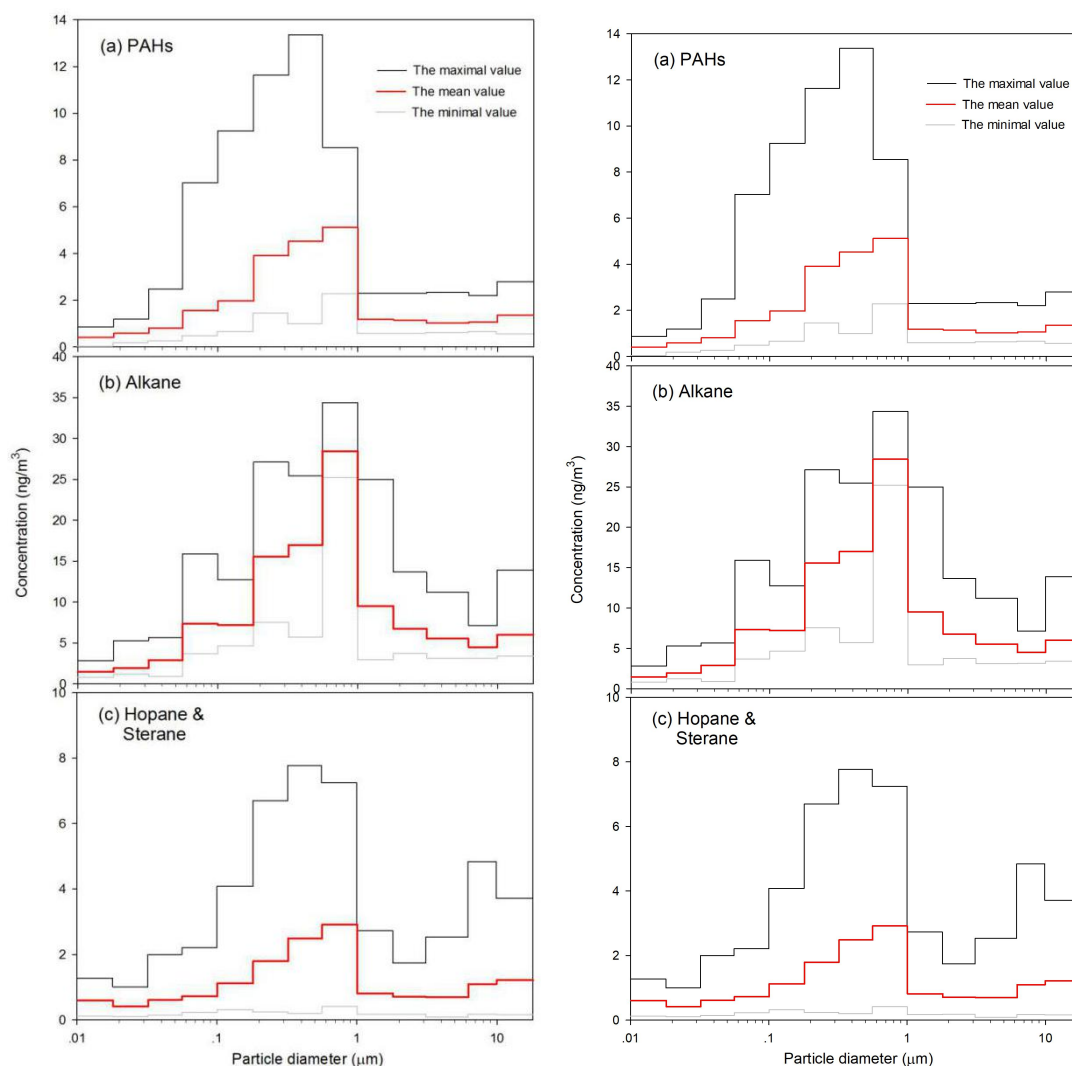
*Response:* The number used in line 215 “Organic matter (OM) was the most abundant component in PM<sub>2.5</sub>, accounting for 18.8–27.8% of the total mass” was our own data which calculated by the measured OC in Jiujiang city. Considering

referee's suggestion, this statement was changed to "Organic matter (OM) was the most abundant component in PM<sub>2.5</sub>, accounting for 18.8–27.8% of the total mass in this study" in lines 237-238 in the revised manuscript.

According to previous researches which studying OM concentrations in Shanghai in YRD area, OM was calculated as  $OM=1.4 \times OC$ , just as "which was estimated to be 1.4 times of OC concentration (Feng et al., 2006; Huang et al., 2014)." in lines 238-239 in the revised manuscript.

**15. Figure 4, the image resolution is poor.**

**Response:** According to referee's suggestion, the Figure 4 was replaced by another figure with high resolution.



**Fig. 4.** Mean-normalized size-specific distribution of NPOCs in the collected PM<sub>2.5</sub> samples (Left for the original figure, Right for the revised figure)

**16. Line 333-335, References should be cited.**

**Response:** According to referee's suggestion, references were cited in this revised manuscript, revised as "Photochemical decay could cause the ambient data to be distributed along a line emanating from the source profile, with

increasing photochemical age (Robinson et al., 2006; Yu et al. 2011).” in lines 355-357 in the revised manuscript.

**17. Line 406, Line 362, Line 400, the abbreviations of the PAH compounds should be given the full expressions.**

**Response:** According to referee’s suggestion, the abbreviations of PAHs were given in full expressions. And the abbreviation,  $P^{\circ}_L$  and  $\Delta H_0$  information for individual NPOCs in Table S1 in the Supplementary Material, was changed to the main manuscript as Table 2.

**Table 2.** Abbreviation,  $P^{\circ}_L$  and  $\Delta H_0$  information for individual NPOCs

Species	Abb.	$P^{\circ}_L$ <sup>a</sup>	$\Delta H_0$ <sup>b</sup>	Species	Abb.	$P^{\circ}_L$	$\Delta H_0$
<b>PAHs</b>				<b>n-Alkanes</b>			
Fluorene	FLO	1.10E-01	84.9	n-Undecane	C <sub>11</sub> H <sub>24</sub>	/	/
Phenanthrene	PHE	2.57E-02	88.9	n-Dodecane	C <sub>12</sub> H <sub>26</sub>	/	/
Anthracene	ANT	1.21E-03	99.7	n-Tridecane	C <sub>13</sub> H <sub>28</sub>	/	/
Fluoranthene	FLU	1.60E-03	98.3	n-Tetradecane	C <sub>14</sub> H <sub>30</sub>	/	/
Pyrene	PYR	7.60E-04	97.9	n-Pentadecane	C <sub>15</sub> H <sub>32</sub>	/	/
Benz[a]anthracene	BaA	3.45E-05	108	n-Hexadecane	C <sub>16</sub> H <sub>34</sub>	/	/
Chrysene	CHR	1.36E-06	118.8	n-Heptadecane	C <sub>17</sub> H <sub>36</sub>	/	/
Benzo[b]fluoranthene	BbF	1.00E-06	119.2	n-Octadecane	C <sub>18</sub> H <sub>38</sub>	/	/
Benzo[j+k]fluoranthene	BkF	4.66E-06	113	n-Nonadecane	C <sub>19</sub> H <sub>40</sub>	/	/
Benzo[a]fluoranthene	BaF	4.66E-05	113	n-Eicosane	C <sub>20</sub> H <sub>42</sub>	/	/
Benzo[e]pyrene	BeP	7.89E-07	117.9	n-Heneicosane	C <sub>21</sub> H <sub>44</sub>	/	/
Indeno[1,2,3-cd]pyrene	IcdP	1.42E-06	124	n-Docosane	C <sub>22</sub> H <sub>46</sub>	3.24E-03	115
Dibenz[a,h]+[a,c]anthracene	DahA	4.93E-09	134.1	n-Tricosane	C <sub>23</sub> H <sub>48</sub>	1.22E-03	120
Benzo[ghi]perylene	BghiP	1.01E-08	129.9	n-Tetracosane	C <sub>24</sub> H <sub>50</sub>	4.66E-04	124
Coronene	COR	3.56E-10	143.2	n-Pentacosane	C <sub>25</sub> H <sub>52</sub>	1.72E-04	129
<b>iso-Alkane</b>				n-Hexacosane	C <sub>26</sub> H <sub>54</sub>	6.59E-05	133
Pristane	C <sub>19</sub> H <sub>40</sub>	/ <sup>c</sup>	/	n-Heptacosane	C <sub>27</sub> H <sub>56</sub>	2.53E-05	137
Phytane	C <sub>20</sub> H <sub>42</sub>	/	/	n-Octacosane	C <sub>28</sub> H <sub>58</sub>	9.42E-06	142
<b>Hopane</b>				n-Nonacosane	C <sub>29</sub> H <sub>60</sub>	3.55E-06	146
$\alpha\beta$ -Nnorhopane	C <sub>29</sub> - $\alpha\beta$ -NOR-H	2.74E-06	126	n-Triacontane	C <sub>30</sub> H <sub>62</sub>	1.32E-06	151
$\alpha\beta$ -Hopane	C <sub>30</sub> - $\alpha\beta$ -H	1.01E-06	130	n-Hentriacontane	C <sub>31</sub> H <sub>64</sub>	4.96E-07	155
$\alpha\beta$ -22R-Homohopane	C <sub>31</sub> - $\alpha\beta$ -R	3.85E-07	134	n-Dotriacontane	C <sub>32</sub> H <sub>66</sub>	1.93E-07	160
ab 22S-Homohopane	C <sub>31</sub> - $\alpha\beta$ -S	3.85E-07	134	n-Tritriacontane	C <sub>33</sub> H <sub>68</sub>	7.09E-08	164
22,29,30-Trisnorhopane	Tm	1.93E-05	117	n-Tetatriacontane	C <sub>34</sub> H <sub>70</sub>	2.63E-08	169
<b>Sterane</b>				n-Pentatriacontane	C <sub>35</sub> H <sub>72</sub>	1.00E-08	173
$\alpha\alpha\alpha$ -20R Cholestane	$\alpha\alpha\alpha$ -20R-C	2.03E-05	121	n-Hexatriacontane	C <sub>36</sub> H <sub>74</sub>	3.75E-09	177
$\alpha\beta\beta$ -20R Cholestane	$\alpha\beta\beta$ -20R-C	/	/	n-Hepatriacontane	C <sub>37</sub> H <sub>76</sub>	1.42E-09	182
$\alpha\beta\beta$ -20R24S-Methylcholestane	$\alpha\beta\beta$ -20R-MEC	7.60E-06	125	n-Octatriacontane	C <sub>38</sub> H <sub>78</sub>	5.37E-10	186
aaa 20R24R-Ethylcholestane	$\alpha\alpha\alpha$ -20R-EC	/	/	n-Nonatriacontane	C <sub>39</sub> H <sub>80</sub>	2.03E-10	191
$\alpha\beta\beta$ -20R24R-Ethylcholestane	$\alpha\beta\beta$ -20R-EC	2.84E-06	130	n-Tetracontane	C <sub>40</sub> H <sub>82</sub>	7.60E-11	195

<sup>a</sup>: pure compound vapor pressure, unit of Pa at 298 K, cited from And and Hanshaw, 2004, Xie et al., 2013;

<sup>b</sup>: vaporization enthalpy, unit of (KJ mol<sup>-1</sup>) at 298 K, cited from Xie et al., 2013, Wang et al., 2016;

<sup>c</sup>: “/” means lack of related data.

**18. Authors should give some reasons for introduced elemental species, inorganic ions and OC/EC into PMF model.**

**Response:** According to referee's suggestion, the reasons for introduced elemental species, inorganic ions and OC/EC into PMF model were added in the revised manuscript. The added statement was "Additionally, chemical constituent of ambient aerosol is an essential step to identifying major sources and quantifying the corresponding contributions to particulate matter. Individual organic tracers, elemental species, inorganic ions and OC/EC have been demonstrated to be able to provide source apportionment of aerosols." in lines 414-416 in the revised manuscript.

**Special thanks to Referee #3 for his/her good comments and his careful reading our manuscript!**

**Reference:**

Ding, L. C., Ke, F., Wang, D. K. W., Dann, T., & Austin, C. C. (2009). A new direct thermal desorption-gc/ms method: organic speciation of ambient particulate matter collected in Golden, BC. *Atmos. Environ.*, 43(32), 4894-4902.

Ho, S. S., Yu, J. Z., Chow, J. C., Zielinska, B., Watson, J. G., & Sit, E. H., et al. (2008). Evaluation of an in-injection port thermal desorption-gas chromatography/mass spectrometry method for analysis of non-polar organic compounds in ambient aerosol samples. *J. Chromatography A*, 1200(2), 217-27.

Ho, S. S., & Yu, J. Z. (2004). In-injection port thermal desorption and subsequent gas chromatography-mass spectrometric analysis of polycyclic aromatic hydrocarbons and n-alkanes in atmospheric aerosol samples. *J. Chromatography A*, 1059(1-2), 121-129.

Ho, S. S. H., Chow, J. C., Watson, J. G., Ng, L. P. T., Kwok, Y., & Ho, K. F. (2011). Precautions for in-injection port thermal desorption-gas chromatography/mass spectrometry (TD-GC/MS) as applied to aerosol filter samples. *Atmos. Environ.*, 45(7), 1491-1496.

Wang, Q., Feng, Y., Huang, X. H. H., Griffith, S. M., Zhang, T., Zhang, Q., Wu, D., and Yu, J. Z. (2016). Non-polar organic compounds as PM<sub>2.5</sub> source tracers: Investigation of their sources and degradation in the Pearl River Delta, China, *J. Geophys. Res. Atmos* 121, 11862-11877.

Yadav, S., Tandon, A., and Attri, A. K. (2013). Characterization of aerosol associated non-polar organic compounds using TD-GC-MS: a four year study from Delhi, India, *J. Hazard. Materials*, 252-253, 29-44.

Yang C., Xu J., Li H. Hong Y. (2016). Temporal variation of PM<sub>2.5</sub> and its relationship with meteorological conditions in Jiujiang City. *Jiangxi Science*, 34(6):790-794. (In Chinese);



**Anonymous Referee #1**

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**This study analyzed the molecular composition of non-polar organic compounds (NPOCs) in PM<sub>2.5</sub> and their size distributions at Jiujiang city. The target NPOCs include n-alkanes, PAHs, and hopanes. Diagnostic ratios and PMF model were applied to the compositional data to evaluate the sources and atmospheric processing of PM<sub>2.5</sub>. In general, this work is well organized and written. However, I still think this work lacks novelty, and would not recommend this manuscript to be accepted for publication at Atmos. Chem. Phys., although a lot of chemical and data analysis work have been done.**

**Response:** To the best of our knowledge, this study was the first one which systematically researched the size-specific distributions (0.01-18 μm), photo-degradation and gas-particle partitioning of NPOCs (PAHs, alkane, hopane and sterane), combined diagnostic ratios of special species and receptor model assessing the effect of partitioning on the source apportionment of PM<sub>2.5</sub> aerosol. The new information on the profiles of PM<sub>2.5</sub>-associated NPOCs, size-specific distributions, effect of gas-particle partitioning to the aerosol identification provided by this study, would help us accurately identify the potential sources of aerosols and then assess the contributions from each source.

**General comments:**

**1. PMF model was utilized to apportion PM<sub>2.5</sub> components to factors/sources. However, the author did not provide any information about the method in the manuscript or supporting information. Which version of PMF model (PMF2 or EPA PMF 5.0) was used for source apportionment? How did the author determine the factor number? How did the author deal with the missing values or measurements below detection limit? Measurement uncertainty was required for PMF input, where were these data from or how were they calculated? Are there any uncertainty analysis related to the PMF modeling? Are the PMF results valid?**

**Response:** Due to the limitation of article length, some detailed description was not added in the original manuscript. According to reviewer's suggestion, the description of PMF analysis would be added in the Section S1 *PMF analysis and uncertainty assessment* of the revised version of Supplementary material (line 27-53). The detailed information was as following:

“Positive matrix factorization (PMF) is considered an advanced algorithm among various receptor models, which has been successfully applied for source identification of environmental pollutants (Han et al., 2014; Besis et al., 2016; Han et al., 2018). PMF has the following advantages: each data point is given an uncertainty-weighting; the factors in PMF

are not necessarily orthogonal to each other and there is no non-negativity constraint with PMF. In the present study, PMF 5.0 (US EPA) was used to apportion the contributions of different sources to PM<sub>2.5</sub> in the atmosphere. The matrix X represents an ambient data set in which i represents the number of samples and j the number of chemical species. The goal of multivariate receptor modeling is to identify a number of sources (p), the species profile (f) of each source and the amount of mass (g) contributed by each source to each individual sample as well as the residuals (e<sub>ij</sub>), as following equation:

$$X_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (S1)$$

The PMF solution minimizes the objective function Q based on these uncertainties (u):

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left[ \frac{X_{ij} \sum_{k=1}^p g_{ik} f_{kj}}{u_{ij}} \right]^2 \quad (S2)$$

The input data files of PMF consist of concentrations and uncertainty matrices, and the uncertainty data were calculated as Equation (S3) as suggested by PMF User Guide. The missing values were represented by average values, while measurements below MDL (method detection limit) were replaced by two times of the corresponding MDL values. The “weak” variables were down-weighted, while “bad” variables were omitted from the analysis process.

$$\begin{cases} \text{Unc}_i = \frac{5}{6} \times \text{MDL}_i & C \leq \text{MDL}_i \\ \text{Unc}_i = \sqrt{(C_i \times \text{ErrorFraction})^2 + (\text{MDL}_i/2)^2} & C > \text{MDL}_i \end{cases} \quad (S3)$$

The model was run 20 times with 25 random seeds to determine the stability of goodness-of-fit values. It is necessary to test different numbers of sources to find the optimal number of sources which produces the most reasonable results. If the number of sources is estimated properly, the theoretical Q value should be approximately the number of degrees of freedom or the total number of data points. Five to eleven factors were examined, and eight factors were found to be the most appropriate and most reasonably interpretable. Q (True) is the goodness-of-fit parameter calculated including all points, while Q (Robust) is the goodness-of-fit parameter calculated excluding points not fit by the model, Q (Robust) and Q (True) were 1,752.4 and 1,812.9, respectively. Additionally, approximately 98% of the residuals calculated by PMF were within the range of -3 to 3, indicating a good fit of simulated results. The factor did not show oblique edges, suggesting there were little rotation for the solution. All these features implied the model simulation result was acceptable.” in Section S1 in the revised Supplementary Materials.

**2. Page 23, line 449-457. This manuscript stated that the PMF<sub>P</sub> and PMF<sub>T</sub> profiles are similar, and should be attributed to the fact that the major NPOCs are enriched in particle phase. This might not apply for factor 6, 7 and 8 (Figure 8f, g and h). The impact of G-P partitioning process will mainly affect the factors highly loaded with low molecular weight species. So the author might need to discuss the impacts of G-P partitioning on these factors.**

*Response:* The lower molecule weight species in gas-particle partitioning were more susceptible to be influenced by ambient temperature, hence light NPOCs show large fugacity from aerosol surfaces. In the present research, factor 8 was recognized as “Light NPOCs” for the characterization of high load of light NPOCs compounds. However, due to PM<sub>2.5</sub> aerosols in this study was mainly conducted in the cold period of high-frequency haze episodes, the resolved factor profiles between PMF<sub>P</sub> and PMF<sub>T</sub> model were similar, even for the light NPOCs factor. The discussion of impact gas-particle partitioning on these factor can be seen detailedly in line 448-456 in the Section of “3.4.3 Assessing impacts of gas-particle partitioning on source apportionment” in this original manuscript.

However, for the factor 6 and 7, namely “Biomass burning” and “Shipping and diesel exhaust”, which were characterized by inorganic salts (Cl<sup>-</sup> with K<sup>+</sup>) and heavy metals (Ni with V), respectively. The tracers for these factors could not be partitioned between phases, despite several light NPOCs species took relative medium to high factor loads, their impacts caused by gas-particle partitioning should be ignored.

**3. From the title, it seems that the manuscript focused on the size distribution, G-P portioning of NPOCs, and the application of NPOCs on source apportionment of PM<sub>2.5</sub>. While this study did not measure the gas-phase NPOCs, the gas-particle portioning is only simulated basing on Pankow’s theory, and could not be validated. As such, it might not be appropriate to put G-P partitioning in the title, or we can say “G-P partitioning simulation”, or “the impacts of G-P portioning on source apportionment”. Size distribution was measured for NPOCs, which should be related to other parts of this manuscript. For example, does the size distribution help to explain the PMF results?**

*Response:* We still think the original “Non-polar organic compounds in aerosols in a typical city of Eastern China: Size distribution, gas-particle partitioning and tracer for PM<sub>2.5</sub> source apportionment” was very proper, for two major reasons. 1). The size-distribution and gas-particle partitioning of NPOCs was really two major research contents in this study. The gaseous phase of NPOCs for the corresponding 13-staged aerosols were not measured in this study, for the season of it is still almost impossible to collect different size-specific particulate and the corresponding gaseous NPOCs simultaneously. In fact, we adopted the classical gas-particle partition model to simulate the abundance of gaseous NPOCs, and explore the particle fraction ( $\phi$ ) of NPOCs with typical organic matter parameters in urban, rural and background areas. Just as PAHs, alkanes, hopanes and steranes could be called as “NPOCs” in this study, though not all kinds of NPOCs species

were analyzed.

2). The NPOCs were used as tracers for the source identification of PM<sub>2.5</sub> by specific specie ratios and receptor model (PMF), but not source apportionment of NPOCs themselves. If the title was changed to “gas-particle partitioning simulation” or “the impacts of gas-particle partitioning on source apportionment”, either it lost the key section of “source identification of PM<sub>2.5</sub>” or it is wrong for generalizing the major research objects for this study.

The size-specific distributions of NPOCs have important influence on their gas-particle partitioning and photo-degradation. Also, combining the characterized species ratios and model extractions, size-specific distributions of NPOCs have relation to aerosols source identifications. The size-distribution of NPOCs was tightly related to the parts of Sections of “Degradation of organics”, “Gas-particle partitioning” and “PMF source apportionment” in this study.

**4. Diagnostic ratios of n-alkanes, PAHs and hopanes were intensively used to evaluate the sources of NPOCs in previous work. The criteria of diagnostic ratios are qualitative and confusing.**

*Response:* Despite diagnostic ratio was relatively a bit empirical and rough when used for the source identification in some cases, it could easily provide useful information in most situations. Additionally, the combined use of diagnostic ratios of NPOCs and PMF model would provide mutual authentication.

**5. Besides the above comments, the lack of enough novelty is the main issue for this work. The size distribution and diagnostic ratios of NPOCs in typical Chinese cities were intensively investigated (Bi et al., 2005; Zhou et al., 2005; Wang et al., 2009a, b, 2011; Duan et al., 2012). The impacts of G-P partitioning of semi-volatile organic compounds (SVOCs) on PMF source apportionment have been observed and validated by Xie et al. (2013, 2014), and the method of using gas + particle phase SVOCs have been intensively applied in PMF source apportionment studies (Gao et al., 2015; Wang et al., 2016; Zhai et al., 2016). Moreover, this work did not provide any new phenomenon or viewpoints that add our knowledge on size distribution or G-P partitioning of NPOCs, or sources apportionment using NPOCs data.**

*Response:* China is suffering severe complex atmospheric pollution, e.g. the persistent heavy haze pollution in Eastern China. Due to the heterogeneous developments of industrial, economic, geomorphic and environmental conditions, cities are facing different environmental pressures and situations. However, researches of NPOCs were currently focused on megacities in relatively developed costal areas in China, leaving most medial cities be ignored. Undoubtedly,

systematically analyzing aerosols bound NPOCs and learning their tracers for the source apportionment of PM<sub>2.5</sub> in a typical medium city, has great academic and practical values without doubt.

NPOCs as one important class of particles were rather chemically stable, which have been reported by numerous researchers, several researchers use them as tracers for PM<sub>2.5</sub> source apportionment. Just as reviewer suggested (Table R2), there were numerous researches about molecular compositions, sized distributions of organic compounds and their effects on G/P partitioning have been published. However, these researches have their unique research interests, showed different focuses to our manuscript (Table R1), e.g. Wang et al., (2011b), Wang et al., (2009) and Wang et al., (2015) analyzed the concentrations of NPOCs and researched their characterizations. Wang et al., (2011a) reported concentrations, characterization and the size specific (0.4-9.0 μm) distributions of n-alkanes, PAHs and hopanes in three different typical sites, they neither evaluated their gas-particle partitioning of these compounds, nor investigated their sources and the corresponding contribution. Xie et al., (2014) evaluated the gas-particle partitioning process of six species of PAHs, twelve species of n-alkanes, hopanes and steranes, explored the partitioning impacts on their source apportionment, and got five NPOCs species profiles (odd alkane, light SVOCs, n-alkane, PAHs and sterane).

This manuscript systematically provides new information on the profiles and characterizations of PM<sub>2.5</sub>-associated NPOCs, evaluated their size-specific distributions and impacts on the gas-particle partitioning, found the effects of gas-particle partitioning and degradation were not apparent o the source apportionment. As best as our knowledge, this is the first research systemically analyzing the characterization, size-specific distribution, gas-particle phase partitioning of NPOCs, and exploring effects of partitioning between tracers for the aerosol source identifications. Based on this manuscript, it will help us to identify the more accurate sources of aerosols and asses the contributions from each source, provide information for further targeted optimized emission control strategies.

**Table R2** Comparison between related studies with this manuscript

ID	Studies	Sampling site	Sampling duration	Analysis method	PAHs	n-Alkanes	Hopanes	Steranes	Size-distribution	Gas-particle partition	Source identification	Remark
①	Wang et al., 2011a; ACP	Baoji city, China ----- Mount Tai, China ----- Okinawa Island, Japan	11-14, Jan. 12-20, Feb. 12-24 Apr. 2008 ----- 22-29 Jun. 2006; 12-24 Jan. 2008 ----- 18 Mar. -12 Apr. 2008	Ultrasonication Extraction+GC/ MS	16 species	C18-C36	5 species	/	TSP; 0.4-9.0 µm, nine stage;	/	/	
②	Wang et al., 2011b; EST	Nanjing city, China	1-17 Jan. 2007 12-14 Oct. 2007	Ultrasonication Extraction+GC/ MS	16 species	C18-C34	/	/	0.4-9.0 µm, nine stage;	/	/	
③	Wang et al., 2009; Tellus, B	The same to study ①	The same to study ①	Ultrasonication Extraction+GC/ MS	/	/	/	/	0.4-9.0 µm, nine stage;	/	/	Mainly about sugar, sugar-alcohol, carboxylic acid
④	Wang et al., 2015; EST	Nanjing city, China	Jul. 2004 - Jan. 2005	Solvent extraction+ GC/MS	/	C18-C36	/	/	/	/	Tracers for source identification	
⑤	Wang et al., 2006; EST	14 Chinese cities	2 days in winter + 2 days in summer, 2003	Ultrasonication Extraction+GC/ MS	18 species	C16-C35	C27-C32	/	/	/	/	Other sugars and so on
⑥	Gao et al., 2015; AE	Guizhou city, China	28 Nov. - 23 Dec. 2009	Ultrasonication Extraction+GC/ MS	13 species	/	4 species	/	/	/	Factor identification + correlation analysis	
⑦	Xie et al., 2014; EST	Denver, USA	Aug. 2012- Jul. 2013	/	6 species	12 species	5 species	5 species	/	/	PMF model	Extracted five species profiles
	This manuscript; ACP	Jiujiang city, China	Sep. - Dec. 2016	TD-GC/MS, without solvent extraction	15 species	30species, C11-C40	5 species	5 species	13 stage, 0.01-18 µm	Partitioning between all these NPOCs	Tracers + PMF model	Extracted 8 factors for PM <sub>2.5</sub> aerosols

We thank Referee #1 for his good suggestions and his/her careful reading our article.

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We tried our best to improve the manuscript and made some changes in the manuscript. These changes will not influence the content and framework of the paper. We appreciate for Editors/ Reviewers' warm work earnestly, and hope that the correction will meet with approval. Once again, thanks very much for your comments and suggestions.

Yours sincerely,

Best regards!

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