A list of responses for comments from editors and reviewers

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 $PM_{2.5}$ source apportionment***" (No. 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 revised manuscript was marked-up with **BLUE** color which we hope meet with approval.

The main corrections in the paper and the responds to the reviewer's comments are as flowing:

Content

Twenty-seven Pages, Three parts.

Suggestions for revision or reasons for rejection

-Reviewer #1

This study investigated non-polar organic compounds including PAHs, alkanes, hopanes and steranes, by using TD-GC/MS technology, in different size-segregated particles from a city (Jiujiang) in eastern China. While the study did provide some information on the characteristics of organic species in a Chinese city, the manuscript in its present form, in my opinion, needs a further clarification and revision before its suitable for publication.

Response: Thanks for reviewer's suggestion, the manuscript has been revised according to editor's and reviewer's suggestions carefully, and the revised section was marked with **BLUE** color in this revised manuscript.

Novelty of the study:

Without doubt that it is valuable to investigate the occurrence of tracer organic species in environment to better reflect the scientific understanding on their sources, impacts and roles in air pollution source apportionment, however, it is still a concern in my opinion that the novelty of this study is very limited. The methodologies including sampling, laboratory analysis and data interpretation are not new. Results and most conclusions are as expected and may be limited to the study area and period. So, what makes such a measurement in a local city novel and scientific that desires for the publication in ACP? (1) What's the implication of this study that is important, may be generated and promote our understanding on the characterization of tracer organics or air pollution? (2) The authors are suggested to rethink about these and add appropriate discussions in the paper before its suitable for publication.

Response: Thanks for reviewer's suggestion. (1). As best as our knowledge, this is the first research systemically analyzing size-specific distribution, photo-degradation and gas-particle partitioning of NPOCs, and evaluating their effects on $PM_{2.5}$ source apportionment. This study was conducted in Jiujiang, a middle scale city in Eastern China, which was more typical than metropolis cities (e.g. Shanghai, Nanjing) for its more complicated emission sources and topography. The $PM_{2.5}$ sources including coal combustion, vehicle emission, oil combustion, ship emission, dust, industrial exhaust, biomass burning, secondary formation and so on. In which, some origins were banned in metropolis cities, such as biomass burning was banned in Shanghai. For the topography, recent researches found that the mountain topography enhanced the pollution degree and lengthened the pollution period, since mountain suited surrounding cities would hinder polluted air mass transportation, condensed under the atmospheric boundary layer, enhancing the atmosphere pollution. The complicated emission sources, typical topography with plain in northern direction, Poyang Lake in the east direction and Mount Lu in the southern direction, all contributed an excellent typical case for researching $PM_{2.5}$ associated NPOCs.

(2). This study found NPOCs distributed in 0.56–1.00 μm were majorly formed from condensation of combustion products. OC/EC promote NPOCs partitioned to particle phase in accumulation mode. Using the predicted gas + measured particle phases NPOCs as input data for PMF analysis, received better factor profiles than particle NPOCs only. These findings will help us accurately identify the potential sources of aerosols and then asses the contributions from each source.

According to reviewer's suggestion, the discussions were revised and further analyzed, the major modifications were as following.

Size-specific distribution. The statement of "This phenomenon could be reasonably explained by that heavy molecular weight PAHs tend to be enriched in smaller particles (< 1.4 μm) (Kleeman et al., 2008), whereas light molecular weight PAHs are speculated to adsorbed onto coarse particles by volatilization and condensation. As discussed above, the heavy molecular weight PAHs accounted for 50.6% of the total PAHs in this study." in lines 327-330 of the original manuscript was revised. The revised description was "<u>This phenomenon could be reasonably explained by that</u>

heavy molecular weight PAHs tend to be enriched in smaller particles (<1.4 μm) (Kleeman et al., 2008), which generally originates from gas-particle transformation, adsorption of gaseous PAHs by condensation or coagulation of combustion products on the surface of preexsiting particles. However, the light molecular weight PAHs are speculated to adsorbed onto coarse particles, mainly originated from resuspension of soil or dust, plant tissue and growing particles from small diameters. As discussed above, the heavy molecular weight PAHs (>4– ring) accounted for 33.7–73.7% (mean of 50.6%) of the total PAHs, and the MDRs values of PAHs species, both confirmed our deduce that the condensation or coagulation of combustion products contributed to the size distribution pattern of PAHs." in lines 336-344 in the revised manuscript.

Additionally, the discussion of "Moreover, our recent research (Han et al., 2018) found that the organic compounds carrier, OC/EC, displayed a unimodal distribution in the fraction of 0.56–1.0 μm among the 13– staged particles. It is also suggested that EC could provide adsorption sites for organic compounds (e.g. NPOCs) due to its large surface area, and has the catalytic properties for redox chemistry reactions. In fact, the relationship between the concentration of NPOCs and the size particles is highly variable. This suggests not only source type but also photodegradation and gas-particle partitioning have great influences on the size-specific distribution of NPOCs, which would be further discussed in Section 3.3 and Section 3.4, respectively." was added in lines 361-366 in the revised manuscript.

Degradation of organics. The statement of "The ratio–ratio plot of IcdP and BghiP normalized by EC from different sites was depicted in Fig. 5a, together with vehicle (diesel and gasoline) exhausts, tunnel and coal burning source profiles. Most of the data points were distributed along a line, which overlapped tunnel, vehicle exhaust and industrial coal, implying ambient hopanes in this city were from vehicle emissions and coal combustion. Wang et al. (2016) reported that vehicle emissions contributed to atmospheric hopanes in four cities in PRD. There were several deviation points at left down corner, and the values were smaller than the values of tunnel and vehicle source profile, indicating mixed influence from traffic origins and degradation. Yu et al. (2011) reported a more apparent liner distributions of data sets measured in Hong Kong and PRD, which can be attributed to their single vehicle source type. In Fig. 5b, most of the data were linearly distributed, implying both biomass burning and vehicle emission contribute to hopanes. Robinson et al. (2006) found hopanes were severely depleted in Pittsburgh, USA, and they attributed this phenomenon to regional air mass transport affecting the oxidation of condensed-phase organic compounds." in lines361-371 of the original manuscript was revised.

The revised description was "Most of the EC normalized IcdP/BghiP data points were distributed along a line (Fig. 5a), implying ambient PAHs underwent photochemical degradation and influenced by vehicle emissions and coal combustion. It was reported that the free ends of C-C scission products of PAHs remain tethered together, which prevent fragmentation and help forming more functional group from the reactions with OH· radical (Hunter et al., 2014). Ultimately, forming the low volatility species which can condense on the particle phase. There were several deviation points at left down corner, and the values were smaller than the values of tunnel and vehicle source profile, indicating mixed influence from traffic origins and degradation. Yu et al. (2011) reported a more apparent liner distributions of data sets measured in Hong Kong and PRD, which can be attributed to their single vehicle source type.

In Fig. 5b, most of the data were linearly distributed, implying the photochemical decay of C29–αβ–NOR/C30–αβ–H in this study. Previous research provided evidences that photochemical oxidation alters the molecular-level composition of hopanes (Robinson et al., 2006). OH· radical was expected to the major atmospheric oxidants, and rudimentary calculations suggest that OH· oxidized hopanes on time scale of a few days which depended on aerosol size. Moreover, the C29–αβ–NOR/C30–αβ–H ratio–ratio plot also suggests both biomass burning and vehicle emission contributed to hopanes concentrations in this study. Robinson et al. (2006) found hopanes were severely depleted in Pittsburgh, USA, and they attributed this phenomenon to regional air mass transport affecting the oxidation of condensed-phase organic compounds." in lines 377-392 in the revised manuscript.

Study site and period:

(3). Why the city is typical in eastern China area? Shall the situation and organic compounds characteristics differ in the studied city from those like Shanghai and Nanjing in east China?

Response: As mentioned above, this study was conducted in Jiujiang, a middle scale city with 4.83 million resident population in 2015, which was the second largest city in Jiangxi Province (there are 6 provinces and one municipality in Eastern China).

For emission sources. The $PM_{2.5}$ sources including coal combustion, vehicle emission, oil combustion, ship emission, dust, industrial exhaust, biomass burning, secondary formation and so on. In which, some origins were banned in metropolis cities, such as biomass burning was banned in Shanghai. There were over 700 thousand motor vehicles in 2015, vehicle emissions were significant; also, Jiujiang Harbor is one huge harbor in the middle of Yangtze River, the ship emission also contributed to the emissions. According to local preliminary statistics, the gross industrial standard coal consumption in Jiujiang amounted to 7.80 million tons in 2015. In contrast to the coastal megalopolis cities (e.g. Shanghai, Nanjing), the domestic coal combustion and biomass combustion were not banned which would emit NPOCs into atmosphere. In Jiujiang, petrochemical industry, which can process approximately five million tons of crude oil per year, is located at the northeast part of the city and in upwind direction. Mount Lu and other surrounding forest provide abundant plant origins for NPOCs.

For the topography, recent researches found that the mountain topography enhanced the pollution degree and lengthened the pollution period, since mountain suited surrounding cities would hinder polluted air mass transportation, condensed under the atmospheric boundary layer, enhancing the atmosphere pollution. The air mass transported from Northern China which brought large amounts of NPOCs, would be accumulated in this city areas for the blocking effect caused by high mountains. The complicated emission sources, typical topography with plain in northern direction, Poyang Lake in the east direction and Mount Lu in the southern direction.

All these mentioned contributed an excellent typical case in this city for researching $PM_{2.5}$ associated NPOCs.

(4). The sampling was from Sep. to Dec. in 2016 (line 102-103), thus this study may be considered as a wintertime study, and results cannot represent a general situation in the city. This info. should be clarified in the title and abstract sections.

Response: The sampling in this study conducted from Sep. to Dec. 2016, mainly represents autumn and early winter time in local climate. Considering reviewer's suggestion, the sampling time in the abstract was clarified, it was modified as "The samples were majorly collected in autumn and winter in a typical city of Eastern China." in line 15-16 of the revised manuscript. Also, other corresponding statement of sampling time was changed, e.g. "Compared with long time investigation, this study was mainly focused in cold season, which would lead to relative high abundance of particle NPOCs with small variation." in line 507-508 of the revised manuscript. The original title "Non-polar organic compounds in aerosols in a typical city of Eastern China: Size distribution, gas-particle partitioning and tracer for $PM_{2.5}$ source apportionment" was changed to "Non-polar organic compounds in autumn and winter aerosols in a typical city of Eastern China: Size distribution and impact of gas-particle partitioning on $PM_{2.5}$ source apportionment" in the revised manuscript.

NPOC analysis using TD-GC/MS:

(5). Line 142-144, Table 2 may be moved to SI as these parameters are mot key part of the study. I'd like to suggest to a new table listing the retention time and quantification ions for each species in the paper. Though the full scan method has been adopted in the study using TD coupled with GC/MS or MSMS technologies, the

accuracy of the quantification is still a big problem. It would be also helpful to provide the mass spectra figure of a representative sample.

Response: Thanks for reviewer's suggestion. The original Table 2 was moved to the Section S2 in the Supplementary Materials. Additionally, the retention time and quantification ions for each NPOCs species was added to this revised table (see **Table R1**). According to reviewer's suggestion, the mass spectra figures of Pyrene, Coronene, n-Docosane and n-Dotriacontane were depicted in the Section S2 in Supplementary Materials, as shown in **Fig. R1**.

Table R1. Abbreviation, P^o_L and ΔH_0, retention time and quantification ion informationinformation for individual NPOCs

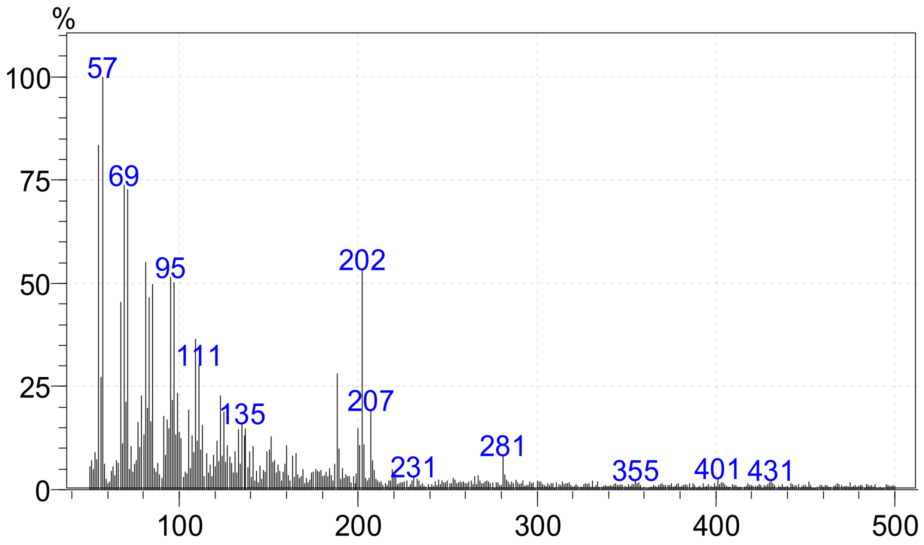
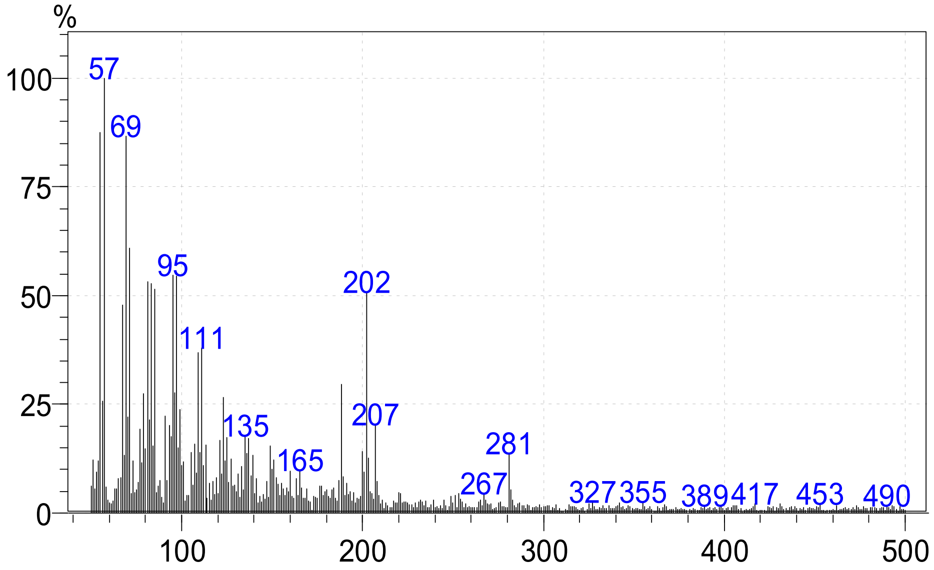
Species	Abb.	P^o_L [a]	ΔH_0 [b]	Base peak (m/z)	Retention Time (RT)
PAHs					
Fluorene	FLO	1.10E–01	84.9	166	19.25
Phenanthrene	PHE	2.57E–02	88.9	178	24.41
Anthracene	ANT	1.21E–03	99.7	178	24.56
Fluoranthene	FLU	1.60E–03	98.3	166	28.03
Pyrene	PYR	7.60E–04	97.9	202	28.66
Benz[a]anthracene	BaA	3.45E–05	108	228	32.30
Chrysene	CHR	1.36E–06	118.8	228	32.41
Benzo[b]fluoranthene	BbF	1.00E–06	119.2	252	35.32
Benzo[j+k]fluoranthene	BkF	4.66E–06	113	252	35.37
Benzo[a]fluoranthene	BaF	4.66E–05	113		36.13
Benzo[e]pyrene	BeP	7.89E–07	117.9	252	36.01
Indeno[1,2,3-cd]pyrene	IcdP	1.42E–06	124	276	39.22
Dibenz[a,h]+[a,c]anthracene	DahA	4.93E–09	134.1	278	39.00
Benzo[ghi]perylene	BghiP	1.01E–08	129.9	276	38.91
Coronene	COR	3.56E–10	143.2	300	28.71
iso–Alkane					
Pristane	$C_{19}H_{40}$	/ [c]	/	57	23.24
Phytane	$C_{20}H_{42}$	/	/	57	24.69
Hopane					
αβ–Nnorhopane	C_{29}–αβ–NOR–H	2.74E–06	126	191	37.77
αβ–Hopane	C_{30}–αβ–H	1.01E–06	130	191	38.54
αβ–22R–Homohopane	C_{31}—αβ–R	3.85E–07	134	191	39.56
ab 22S–Homohopane	C_{31}—αβ–S	3.85E–07	134	191	39.70
22,29,30–Trisnorhopane	Tm	1.93E–05	117	191	36.63
Sterane					

ααα–20R Cholestane	ααα–20R–C	2.03E–05	121	217	37.29
αββ–20R Cholestane	αββ–20R–C	/	/	218	37.66
αββ–20R24S–Methylcholestane	αββ–20R–MEC	7.60E–06	125	218	36.58
aaa 20R24R–Ethylcholestane	ααα–20R–EC	/	/	217	37.29
αββ–20R24R–Ethylcholestane	αββ–20R–EC	2.84E–06	130	218	37.66
n–Alkanes					
n–Undecane	$C_{11}H_{24}$	/	/	57	12.39
n–Dodecane	$C_{12}H_{26}$	/	/	57	13.92
n–Tridecane	$C_{13}H_{28}$	/	/	57	16.10
n–Tetradecane	$C_{14}H_{30}$	/	/	57	18.15
n–Pentadecane	$C_{15}H_{32}$	/	/	57	20.26
n–Hexadecane	$C_{16}H_{34}$	/	/	57	21.63
n–Heptadecane	$C_{17}H_{36}$	/	/	57	23.15
n–Octadecane	$C_{18}H_{38}$	/	/	57	24.55
n–Nonadecane	$C_{19}H_{40}$	/	/	57	25.87
n–Eicosane	$C_{20}H_{42}$	/	/	57	27.12
n–Heneicosane	$C_{21}H_{44}$	/	/	57	28.33
n–Docosane	$C_{22}H_{46}$	3.24E–03	115	57	29.43
n–Tricosane	$C_{23}H_{48}$	1.22E–03	120	57	30.61
n–Tetracosane	$C_{24}H_{50}$	4.66E–04	124	57	31.55
n–Pentacosane	$C_{25}H_{52}$	1.72E–04	129	57	32.43
n–Hexacosane	$C_{26}H_{54}$	6.59E–05	133	57	33.09
n–Heptacosane	$C_{27}H_{56}$	2.53E–05	137	57	33.36
n–Octacosane	$C_{28}H_{58}$	9.42E–06	142	57	33.50
n–Nonacosane	$C_{29}H_{60}$	3.55E–06	146	57	35.47
n–Triacontane	$C_{30}H_{62}$	1.32E–06	151	57	37.31
n–Hentriacontane	$C_{31}H_{64}$	4.96E–07	155	57	39.24
n–Dotriacontane	$C_{32}H_{66}$	1.93E–07	160	57	37.66
n–Tritriacontane	$C_{33}H_{68}$	7.09E–08	164	57	40.22
n–Tetratriacontane	$C_{34}H_{70}$	2.63E–08	169	57	38.75
n–Pentatriacontane	$C_{35}H_{72}$	1.00E–08	173	57	40.21
n–Hexatriacontane	$C_{36}H_{74}$	3.75E–09	177	57	41.33
n–Hepatriacontane	$C_{37}H_{76}$	1.42E–09	182	57	42.82
n–Octatriacontane	$C_{38}H_{78}$	5.37E–10	186	57	43.55
n–Nonatriacontane	$C_{39}H_{80}$	2.03E–10	191	57	45.13
n–Tetracontane	$C_{40}H_{82}$	7.60E–11	195	57	46.21

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

[b]: vaporization enthalpy, unit of (KJ mol^{-1}) at 298 K, cited from Xie et al., 2013, Wang et al., 2016;

[c]: "/" means lack of related data.

(a) Pyrene	
(b) Coronene	

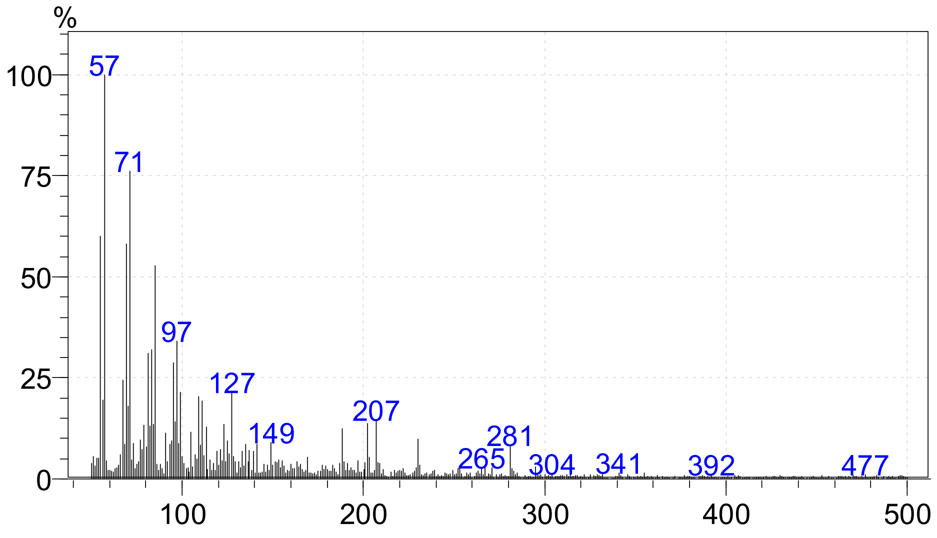
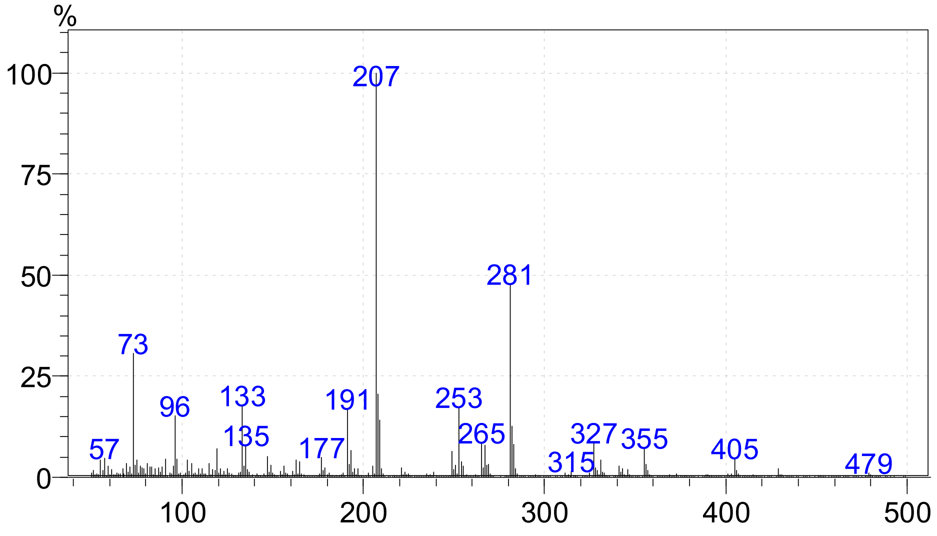
(c) n－Docosane	
(d) n－Dotriacontane	

Fig. R1. The mass spectra of selected NPOCs (a for Pyrene, b for Coronene, c for n－Docosane, d for n－Dotriacontane)

(6). Line 178-is the spiked solution from the standard chemicals or the SRM (line 166-SRM particles)?

QA/QC-are there any blank filters from the field sampling?

Response: The statement of spiked solution in original line 178 referred to the Standard Reference Materials (SRM), including SRM 2260A, SRM 1494 and SRM 2266. Considering reviewer's suggestion, this depiction was changed to "The analytical recovery was calculated via spiking a known amount of the SRM solution to blank filter" in line

176-177 of the revised manuscript.

In the Quality assurance and quality control section, not only blank filters from filed sampling, but also transport blank and laboratory blank filters were analyzed in this study. It was depicted as "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 159-162 in the revised manuscript.

Gas-particle partitioning

(7). Without a direct measurement of gaseous organics, the whole discussion and interpretation on the gas-particle partitioning in this paper is very weak and inconclusive. There are also many other gas-particle partitioning models, of which there are very rough estimated numbers for some key parameter. The authors may still keep a small part of discussion on the partitioning and its impacts on the PMF results, however, it is not suggested to highlight this in the title and a long paragraph in the results/discussion part. Those only increased the length of the paper but not the depth and significance of the study, and to some extent have a negative impact of the manuscript organization.

Response: Due to the high subcooled liquid vapor pressure or octanol-air partitioning coefficient value, heavy NPOCs, e.g. long chain n-alkane (>27 C), PAHs (molecular weight >252), hopane and sterane are mostly in particle phase. There was limited study about gaseous NPOCs have been reported. When the measured gaseous phase NPOCs data were not available, the predicted gaseous NPOCs from gas-particle model provides as a good substitution.

Among different gas-particle partitioning models, the one developed by Pankow (1994 a,b) was the most widely adopted for the partitioning of semi-volatile organic compounds (SVOCs), e.g. NPOCs, PCBs, PBDEs and divalent mercury. This gas-particle partitioning model took account of major affecting factors, such as molecular weight of the absorbing OM phase, the mole fraction scale activity coefficient (ζ) of each compound in the absorbing OM phase, vapor pressure of each pure compound ($P^{o}L$), ambient temperature (T) and ideal gas constant (R).

In this study, only particle associated NPOCs were measured, without measured gas data to explore gas-particle partitioning process would inevitably bring some uncertainties. However, we do not think the whole discussion was inconclusive, since numerous researches suggested that this partitioning model predicted similar gaseous values comparable to the measured ones. For example, Xie et al. (2014) found the factor contributions were consistent from both the “measured total” and “predicted total” SVOCs, both of which reduced the influence from gas-particle partitioning.

Considering reviewer’s suggestion, the discussion on the partitioning of NPOCs was shortened, and it was not highlighted in the abstract and conclusions. For example, the statement of “Gas-particle partitioning model showed that the particle-phase fraction (φ) of light molecular weight NPOCs ranged from 2.4% to 62.5%, while that of heavy NPOCs accounted for more than 90.0%.” in the original abstract, “Gas-particle partitioning model showed that the calculated particle-phase fraction (φ) of the light molecular weight NPOCs ranged between 2.4% to 62.5%, while those of heavy PAHs, long chain n-alkanes, hopanes and steranes were high (>90.0%).” in the original conclusions, were deleted in the revised manuscript. The words of section “3.4.1 Gas-particle partitioning” (387 words) was shortened to ~a half of the original one, and the figure 7 was also depleted in the revised manuscript.

Source apportionment.

(8). The limitations in those apportionment methods including ratios as well as PMF have been widely recognized, however, these are still widely used nowadays. The limitations are briefly mentioned in some parts (lines 278-280). It is necessary and strongly suggested to have a separate section discussing the limitation of the study including

sampling periods and methods used in source apportionment discussion.

Response: According to reviewer's suggestion, the limitations of PMF model, sampling time and gas-particle partitioning effect were added, "3.5 Limitation and implication" in lines 494-511 in the revised manuscript. It was depicted as "In this work, we confirmed that using the total (gas+ particle) NPOCs as input data for receptor model provides a better source apportionment than using only particle phase. However, the predicted gas NPOCs from gas-particle partitioning may bring some uncertainty. For example, the partitioning process are strongly affected by the particle properties, e.g. particle size, organic carbon compounds and the prevailing ambient temperature. For size-distribution, the $PM_{2.5}$ associated NPOCs in the 0.56–1.00 μm fraction were the most abundant, our recent study also found OC was primarily distributed in this fraction (Han et al., 2018). Abundant OC would adsorb/absorb large amounts of NPOCs, resulting particle bound NPOCs concentration increasing in this particle size.

Low temperature promotes NPOCs adsorbing/absorbing onto aerosols, while photochemical degradation of NPOCs is relatively weak in cold season. Moreover, photochemical reactions would reduce the abundances of organic marker depend on species, significantly altering the relative contribution of different sources extracted by liner source inversion. Compared with long time investigation, this study was mainly focused in cold season, which would lead to relative high abundance of particle NPOCs with small variation.

For PMF model, it has limit that could not identify potential source without preexisting tracer. Also, the relative small number of measurements might lead to some uncertainty in source apportionment. In the future, more source tracers data need to be included for the calculation of potential contributions."

Specific ones:

(9). Title- as mentioned "gas-particle partitioning" is not suggested to be highlighted here. Also "tracers for $PM_{2.5}$

source apportionment" is a little confusing to me. The study did not investigate or evaluate the use of specific trace organics in $PM_{2.5}$ source apportionment. What they did is the use of these tracers, together with ions, to run PMF and to identify potential sources of $PM_{2.5}$. Strongly suggest the authors to rethink the title for this manuscript.

Response: Considering reviewer's suggestion, the original title of "Non-polar organic compounds in aerosols in a typical city of Eastern China: Size distribution, gas-particle partitioning and tracer for $PM_{2.5}$ source apportionment", was revised as "Non-polar organic compounds in autumn and winter aerosols in a typical city of Eastern China: Size distribution and impact of gas-particle partitioning on $PM_{2.5}$ source apportionment" in the revised manuscript.

(10). Line 18-19, suggest to add the quantitative percentages, and the implication of different size distributions between PAHs and hopanes, - sources?

Response: Thanks for reviewer's suggestion, the original description of " the middle chain length n–alkanes (C_{25}–C_{34}) were the most abundant in n-alkanes. PAHs and n-alkanes were majorly distributed in 0.56–1.00 μm fraction. ∑(hopanes+steranes) were associated with the 0.32–1.00 μm fraction." was changed to "the middle chain length n–alkanes (C_{25}–C_{34}) were the most abundant (72.3%) in n-alkanes. PAHs and n-alkanes were majorly distributed in 0.56–1.00 μm fraction, while ∑(hopanes+steranes) were associated with the 0.32–1.00 μm fraction, suggesting condensation of combustion products was their important origins." in line 18-20 in the revised manuscript.

(11). Lines 23-24, no new information added to the current knowledge as this trend has been widely recognized by the researches in this area. Also high uncertainties in these quantitative numbers as this is from a simple model estimation. Suggest to remove.

Response: According to reviewer's suggestion, the statement of "Gas-particle partitioning model showed that the particle-phase fraction (φ) of light molecular weight NPOCs ranged from 2.4% to 62.5%, while that of heavy NPOCs accounted for more than 90.0%. ” was deleted in the revised manuscript.

(12). Line 109- “Han et al., 2018” is missing in the reference section

Response: Thanks for reviewer’s suggestion, the reference “Han, D., Zhang, J., Hu, Z., Ma, Y., Duan, Y., Han, Y., and Cheng J.: Particulate mercury in ambient air in Shanghai, China: Size-specific distribution, gas-particle partitioning, and association with carbonaceous composition. Environ. Pollut., 238, 543-553, 2018.” was added in lines 547-549 in the Section of Reference of the revised manuscript.

(13). Lines 231-232, provide references

Response: The references were added in lines 231-232 of the revised manuscript, revised as “In this study, c = 17.2 Pa cm^{-1}, and θ is 1.1×10^{-5}, 1.5×10^{-6} and 4.2×10^{-7} (cm^2 cm^{-3}) for urban area, rural area and background, respectively (And and Hanshaw, 2004; Xie et al., 2013).”

(14). Line 236, is this a daily average or seasonal?

Response: The average value of $PM_{2.5}$ was daily average concentration, it was revised as “The daily average $PM_{2.5}$ concentration in all sampling sites ” in line 236 in the revised manuscript.

(15). Line 237-238, the sentence “which was estimated…” may be moved to line 237 after the phase “organic matter (OM)”

Response: Thanks for reviewer’s suggestion, this statement was revised as “Organic matter (OM) was estimated to be 1.4 times of OC concentration (Feng et al., 2006;Huang et al., 2014), which was the most abundant component in $PM_{2.5}$, accounting for 18.8–27.8% of the total mass in this study. ” in lines 237-239 in the revised manuscript.

(16). Line 242, “daily average concentration’

Response: This statement of “The total concentrations ranged from” changed to “Their daily average concentrations ranged from ” in line 242 in the revised manuscript.

(17). Lines 243-244, what’re the quantitative percentages of these studies? It is interesting but also a little strange that the results were comparable in a fast-developed area (PRD), the marine aerosol (south China sea study), and the present city in inland. Please provide results and study periods in these literature and discuss if applicable.

Response: The quantitative percentages of NPOCs to OM in PRD South China Sea were 0.1–4.2% and 0.8–1.7%, respectively. Considering reviewer’s suggestion, this description was revised as “This was consistent with the measurement results (0.1–4.2%) of NPOCs in Pearl River Delta (PRD, China) (Wang et al., 2016), over a two-year period from 2011 to 2012. Similarly, Zhao et al., (2016) found that the NPOCs varied from 19.8 to 288.2 ng m^{-3}, accounted for 0.8–1.7% of OM in South China Sea from Sep. to Oct. 2013.” in lines 243-246 in the revised manuscript.

(18). Lines 243-251, when comparing to literature data, it is important and necessary to clarify the study period and year of those in literature. Some data in Table 3 are the annual average, while some are the monthly or maybe daily average. A simple comparison, to the wintertime study in this paper, and the high/low contamination conclusion here is not acceptable.

Response: According to reviewer’s suggestion, the statement in lines 248-251 of the original manuscript, was modified by adding detailed information of each research in lines 250-260 in the revised manuscript. The revised description was “When comparing with other NPOCs measurements in China, Li et al. (2013) reported a comparable level that the daily concentration of n-alkanes, PAHs and hopanes were 97.9, 13.5 and 21.5 ng m^{-3} in Hong Kong in winter, respectively. Xu et al. (2013) measured mean n-alkanes, PAHs concentrations of 48.1±20.8, 19.0±17.5 ng m^{-3} in urban Guangzhou during 16th Asian Games, while Feng et al. (2006) found that the daily concentrations of n-alkanes, PAHs ranged of 32.9–342.9 and 7.8–151.1 ng m^{-3} in urban Shanghai in 2002–2003, respectively. Thus, this study finds generally higher $PM_{2.5}$ associated NPOCs concentrations measured in Jiujiang compared to other measurements, which may be due to this study was mainly conducted in cold season when severe atmospheric pollution episodes (including haze) frequently occurred. In addition, aerosols and NPOCs would transported to Jiujiang from Northern China where has significant amounts of coal burned and industries, through long range transport of air mass (Han et al., 2018). However, the annual average concentrations of PM_{10} bound n-alkanes and PAHs in Delhi, India (Yadav et al., 2013) were 4.0 and 8.2 times higher than that in Jiujiang, respectively. ”

(19). Figure 2- is the left circle $PM_{2.5}$- inconsistent with the legend. Only 5 site results? The area of those might be adjusted to reflect the mass concentration. Also, please pay attention to the significance figures. Did the measurement technology provide an accuracy of 0.01 µg/m3?

Response: Thanks for reviewer’s suggestion, this original figure (**Fig. R2**) was modified by rearranging the legends to match $PM_{2.5}$ and NPOCs constitutes, changing the accuracy of the measured abundance. The modified figure was depicted as **Fig. R3**.

The $PM_{2.5}$ associated NPOCs were measured at five sites synchronously, including XY, SL, JJ, SH and WQ sites. However, the 13-staged size-specific aerosol bound NPOCs were measured at EM site.

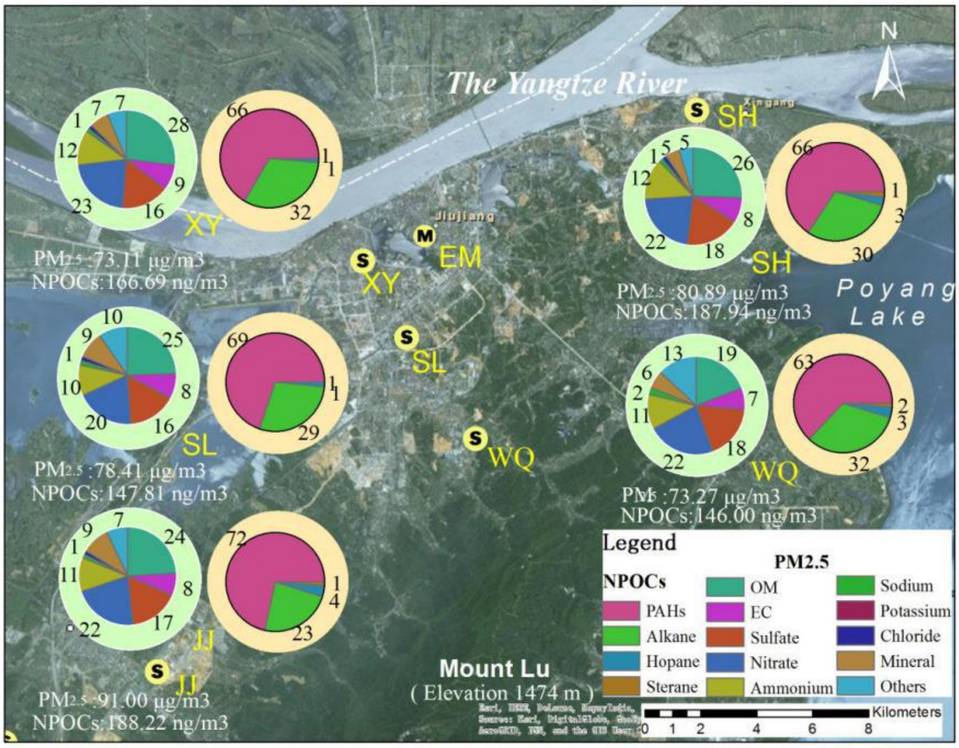


Fig. R2. Spatial distributions of NPOCs and $PM_{2.5}$ in Jiujiang city

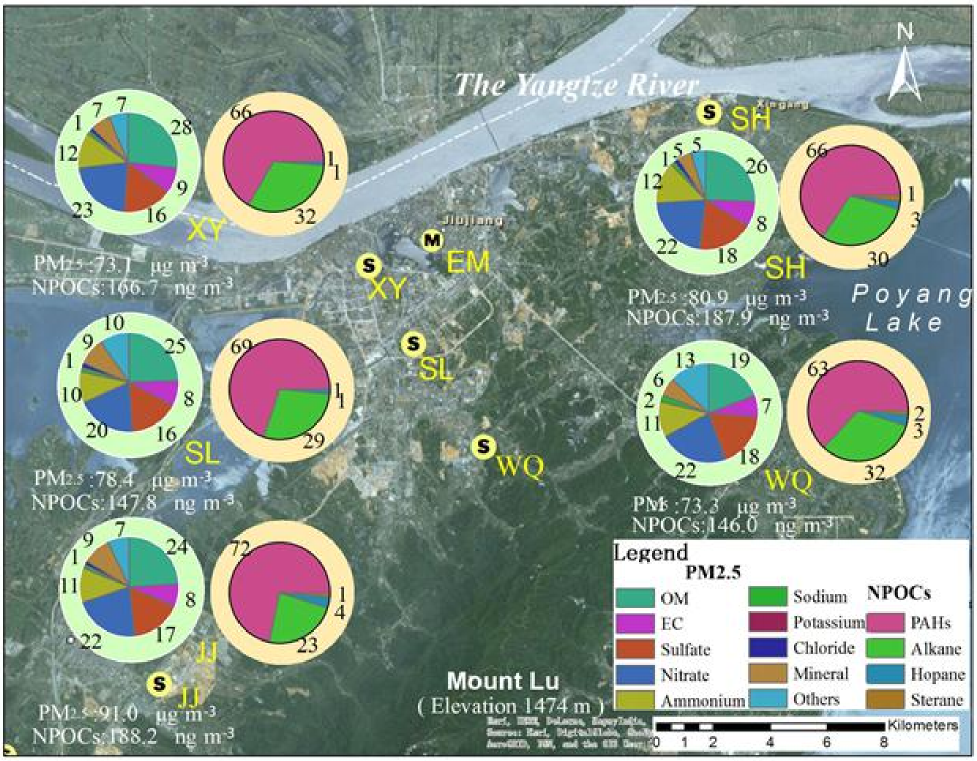


Fig. R3. The percentiles of NPOCs and $PM_{2.5}$ constituents in five sampling sites in Jiujiang city

(20). Line 307- "emissions were similar", or the alkanes are more likely homogeneously distributed across

different sites?

Response: According to reviewer's suggestion, this description was changed to "This small fluctuations may suggest that n-alkanes emissions were similar across different sampling sites, displayed a relative homogeneous distribution in Jiujiang." in lines 316-318 in the revised manuscript.

(21). Line 326, the peak in "9.9-18 µm" is very tiny

Response: The peak value of PAHs in 9.9-18.0 µm was much lower than that in 0.56-1.0 µm, but the peak values of this coarse mode were enhanced for n-alkanes, hopans & steranes, as depicted in Fig 4 in the revised manuscript.

(22). Figure 5- suggest to find another way to improve the presentation of these data. for example, maybe monitoring data from the present study can be in the same white or black sign as spatial distribution is not discussed here.

Response: The original figure (marked as **Fig. R4**) was revised by changing the monitor data to black sign in the revised manuscript, according reviewer's suggestion, and the revised figure was depicted in **Fig. R5**.

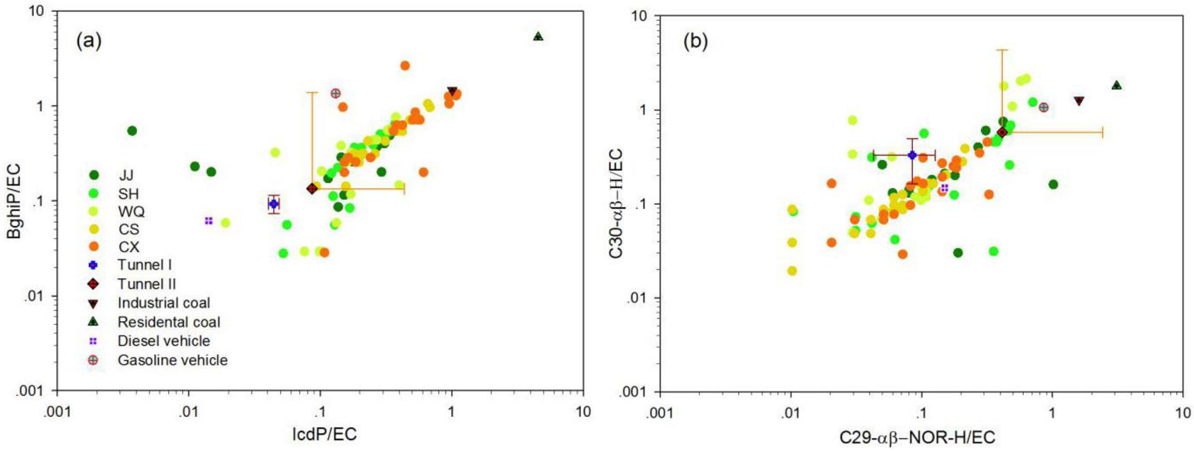


Fig. R4. Ratio–ratio plots of two pairs of characterized species (IcdP/BghiP and C29–αβ–NOR–H/C30–αβ–H) normalized by EC and published source profiles. (Tunnel I: Yu et al., 2011; Tunnel II: He et al., 2009; Residental coal: Zhang et al., 2008; Industrial coal: Zhang et al., 2008; Diesel vehicle: Fraser et al., 2002; Gasoline vehicle: Fraser et al., 2002)

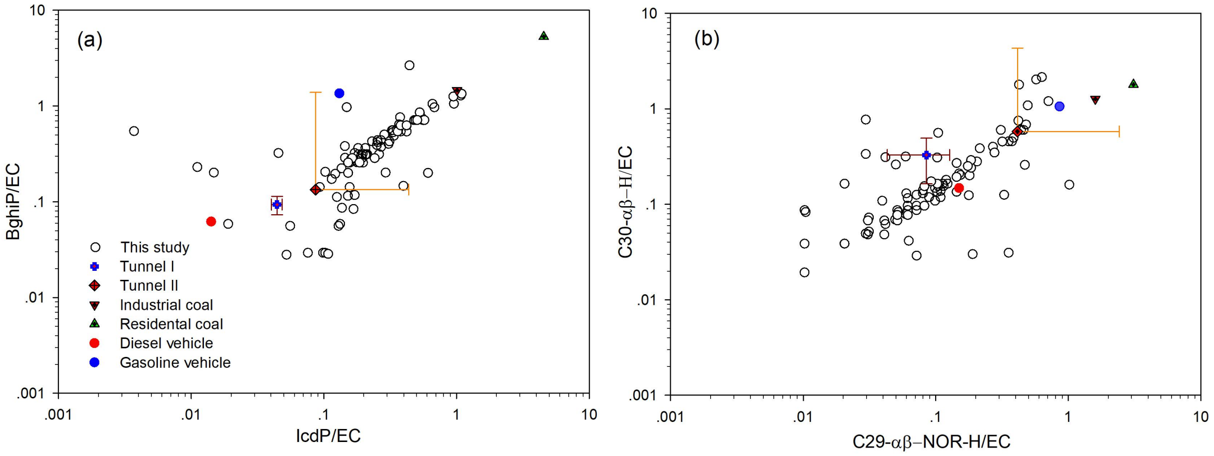


Fig. R5. Ratio–ratio plots of two pairs of characterized species (IcdP/BghiP and C29–αβ–NOR–H/C30–αβ–H) normalized by EC and published source profiles. **(**Tunnel I: Yu et al., 2011; Tunnel II: He et al., 2009; Residental coal: Zhang et al., 2008; Industrial coal: Zhang et al., 2008; Diesel vehicle: Fraser et al., 2002; Gasoline vehicle: Fraser et al., 2002**)**

Special thanks to you for your careful reading and good comments!

-Reviewer 2

This study measured the concentrations of non-polar organic compounds (NPOCs) in $PM_{2.5}$ and size resolved particles. Gas/particle portioning of the target compounds were also analyzed and considered during the source apportionment of $PM_{2.5}$. In general, this work is well organized and written. I would recommend this work to be accepted after minor revision.

Response: Thanks for reviewer's suggestion, the manuscript has been revised according to reviewer's suggestion carefully, and the revised section was marked with **BLUE** color in this revised manuscript.

Minor revision.

(1). Page 6. Table 2

I would suggest the author to leave this table in supporting information.

Response: According to reviewer's suggestion, this table was moved to Section S2 of the Supplementary Materials.

(2). The method for source apportionment was provided in supporting information, would the author please mention it somewhere in the manuscript?

Response: Considering reviewer's suggestion, the PMF model was mentioned in the revised manuscript, depicted as "To explore the impact of gas-particle partitioning on $PM_{2.5}$ source apportionment, both single particle phase and the total (gas+ particle) NPOCs were incorporated with elemental species, inorganic ions and OC/EC, used as input data for receptor model PMF (detailed description about PMF model could be seen in Section 2 of Supplementary Materials)." in lines 423-426 of the revised manuscript.

(3). Page 20, lines 414-420, and page 22 figure 8

Where are the elements and ions data from? If they are reported for the first time in this work, please provide the measurement method and QA/QC procedure. If not, please provide some references.

Response: The measurement methods of OC/EC, elements and ions data were described in the section of "2.3 Determination of OC/EC and other constituents". According to reviewer's suggestion, the Quality Assurance and Quality Control was added in lines 146-156 in the revised manuscript.

The revised statement of their measurement was "OC and EC were analyzed (a round punch of 0.538 cm^2) using the thermal–optical– transmittance (TOT) method (NIOSH protocol, Desert Research Institute, USA) (Han et al., 2018). The instrument included a temperature- and atmosphere-controlled oven and a laser of 680 nm wavelength to generate an operational EC/OC split. The instrument was heated stepwise from start to 250 ºC (60 s), 500 ºC (60 s), 650 ºC (60 s) and finally 850 ºC (90 s) in the helium atmosphere for OC volatilization, and from start to 550 ºC (45 s), 650 ºC (60 s), 750 ºC (60 s) and finally 850 ºC (80 s) in the helium atmosphere containing 2% oxygen for EC oxidation.

Elemental compositions, including Na, K, Ca, Mg, P, Fe, Ti, Al, Pb, Cu and Zn, were determined by energy dispersive X-ray fluorescence (ED-XRF) spectrometry (Epsilon 5, Netherlands). Water soluble inorganic ions, including cations (Na^+, K^+, Mg^{2+}, Ca^{2+}, NH_4^+) and anions (Cl^-, SO_4^{2-} and NO_3^-, NO_2^-), were detected by ion chromatography (IC, ISC-90, Dionex, USA). The detailed experimental procedure of OC/EC, elemental composition, inorganic ions analysis could be found in Li et al. (2017) and Han et al. (2018)."

(4). Page 23, lines 484-486.

Please add some references for this statement.

Response: Considering reviewer's suggestion, the references were added in lines 502-504 in the revised manuscript, the revised statement was "While gas phase oxidation reaction is much faster than heterogeneous reactions in aerosol surface, since the uptake of heterogeneous oxidant is diffusion-limited (Robinson et al., 2006;May et al., 2012)."

Special thanks to you for your careful reading and good comments!

-Other major modifications

The total words of the whole original manuscript was 10228, while the revised manuscript was 10155 words totally.

Abstract.

The abstract was shortened from 279 words to 256 words, and the statement of "This study provides new information on the profiles of $PM_{2.5}$-associated NPOCs, size-specific distributions, photo-degradation and their gas-particle partitioning. This will help us accurately identify the potential sources of aerosols and then asses the contributions from each source.", was revised to "These findings highlight the emissions from different aerosols associated NPOCs origins, caused different size-specific distributions, photo-degradation and gas-particle partitioning, which further affect $PM_{2.5}$ source apportionment. Considering these effects on organic tracers will help us accurately identify the potential sources of aerosols and then asses the contributions from each source." in lines 25-28 in the revised manuscript.

Abbreviations of the PAHs, hopanes and steranes compounds.

The abbreviations of PAHs, hopanes and steranes in the main manuscript were given in full expressions, e.g. "(6) MDRs for PAHs source apportionment include ANT/PHE atio, PYR/FLU ratio, IcdP/BghiP ratios" was revised to "(6) MDRs for PAHs source apportionment include ANT/PHE (Anthracene/Phenanthrene) ratio, PYR/FLU (Pyrene/Fluoranthene) ratio, IcdP/BghiP (Indeno[1,2,3-cd]pyrene/Benzo[ghi]perylene) ratios" in lines 202-203 in the revised manuscript.

"BbF (5.7 ng m^{-3}) was the most abundant PAH species, followed by BaA (5.6 ng m^{-3}) and BaP (4.2 ng m^{-3})" was changed as "BbF (Benzo[b]fluoranthene, 5.7 ng m^{-3}) was the most abundant PAH species, followed by BaA (Benz[a]anthracene, 5.6 ng m^{-3}) and BaP (Benzo[e]pyrene, 4.2 ng m^{-3})" in lines 271-272 in the revised manuscript.

"The predominant hopane analogs were C_{30}–αβ–H, C_{31}–αβ– and C_{29}–αβ– NOR–H, with concentrations of 1.2±1.3, 0.8±0.7 and 0.8±0.9 ng m^{-3}, respectively." was modified as "The predominant hopane analogs were C_{30}–αβ–H (αβ–Hopane), C_{31}–αβ–S (ab 22S–Homohopane) and C_{29}–αβ– NOR–H (αβ–Nnorhopane), with concentrations of 1.2±1.3, 0.8±0.7 and 0.8±0.9 ng m^{-3}, respectively." in lines 325-326 in the revised manuscript.

Reference

Han, D., Zhang, J., Hu, Z., Ma, Y., Duan, Y., Han, Y., and Cheng J.: Particulate mercury in ambient air in Shanghai, China: Size-specific distribution, gas-particle partitioning, and association with carbonaceous composition. Environ. Pollut., 238, 543-553, 2018.

Hunter, J. F., Carrasquillo, A. J., Daumit, K. E., Kroll, J. H.: Secondary organic aerosol formation from acyclic, monocyclic, and polycyclic alkanes. Environ. Sci. Technol., 48(17), 10227-34, 2014.

Wang, Q., Feng, Y., Huang, X. H. H., Griffith, S. M., Zhang, T., Zhang, Q., Wu, D., and Yu, J. Z.: Non-polar organic compounds as PM2.5 source tracers: Investigation of their sources and degradation in the Pearl River Delta, China, J. Geophy. Res. Atmos, 2016.

Xie, M., Barsanti, K. C., Hannigan, M. P., Dutton, S. J., and Vedal, S.: Positive matrix factorization of PM2.5 - eliminating the effects of gas/particle partitioning of semivolatile organic compounds, Atmos. Chemi. Phys., 13, 7381, 2013.

Xie, M., Hannigan, M.P., Barsanti, K.C.: Impact of Gas/Particle Partitioning of Semivolatile Organic Compounds on Source Apportionment with Positive Matrix Factorization. Environ. Sci. Technol. 48, 9053-9060, 2014.

Xie, Y., Ye, X., Ma, Z., Tao, Y., Wang, R., Zhang, C., Yang, X., Chen, J., and Chen, H.: Insight into winter haze formation mechanisms based on aerosol hygroscopicity and effective density measurements, Atmos. Chemi. Phys., 17, 7277-7290, 2017.

Yu, J. Z., Huang, X. H., Ho, S. S., and Bian, Q.: Nonpolar organic compounds in fine particles: quantification by thermal desorption-GC/MS and evidence for their significant oxidation in ambient aerosols in Hong Kong, Analytical Bioanalytical Chem., 401, 3125-3139, 2011.

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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