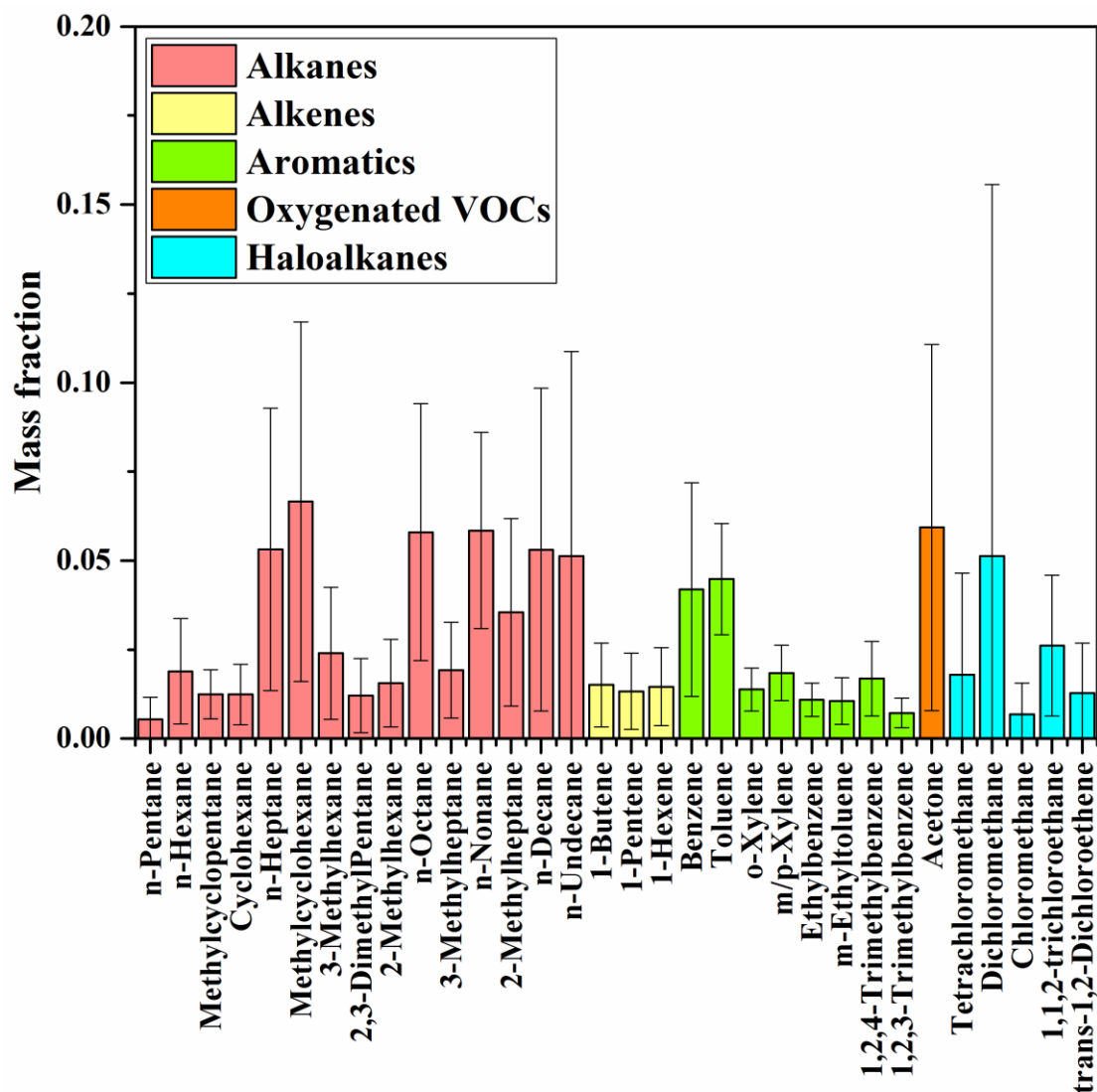


Response to Referee #1

- Figure 2: the y-axis is labelled "mass percentage", but the figures are mass fractions, not mass percentages.

Response: Thanks for the careful review. The label of Y-axis in Figure 2 has been revised accordingly.

Revision in manuscript:



- The corresponding tabulated data in Table 3 have far too many significant figures. I suggest that 2 decimal places at the most would better reflect the uncertainties in the measurements

Response: Thanks for the helpful suggestion. We have revised the decimal places of data in Table.

Revision in manuscript:

Table3: Top 32 VOC species mass percentage from 16 container ships

Compounds	Percentage	Standard deviation	Compounds	Percentage	Standard deviation
n-Pentane	0.54	0.63	1-Pentene	1.33	1.07
n-Hexane	1.89	1.48	1-Hexene	1.46	1.10
Methylcyclopentane	1.24	0.70	Benzene	4.20	3.00
Cyclohexane	1.24	0.85	Toluene	4.48	1.57
n-Heptane	5.31	3.97	o-Xylene	1.38	0.60
Methylcyclohexane	6.66	5.04	m/p-Xylene	1.84	0.78
3-Methylhexane	2.40	1.86	Ethylbenzene	1.10	0.47
2,3-DimethylPentane	1.21	1.04	m-Ethyltoluene	1.06	0.65
2-Methylhexane	1.56	1.23	1,2,4-Trimethylbenzene	1.68	1.05
n-Octane	5.80	3.61	1,2,3-Trimethylbenzene	0.72	0.41
3-Methylheptane	1.99	1.34	Acetone	5.93	5.14
n-Nonane	5.84	2.76	CCl ₄	1.79	2.85
2-Methylheptane	3.55	2.63	CH ₂ Cl ₂	5.13	10.42
n-Decane	5.31	4.53	CH ₃ Cl	0.68	0.87
n-Undecane	5.13	5.74	1,1,2-trichloroethane	2.61	1.98
1-Butene	1.51	1.18	trans-1,2-Dichloroethene	1.27	1.40

Response to Referee #2

1. While compounds with higher carbon numbers are more relevant to secondary organic aerosols, how about the effect of lower-carbon compounds on the ozone formation potential? The effects of neglecting lower-carbon compounds on calculated SOA yields and especially OFP should have been evaluated quantitatively.

Response: *Yes, we should admit that the absence of lower-carbon compounds in GC-MS quantification has impact on the calculated results of ozone forming potential. According to Gentner's research (Gentner et al., 2012), hydrocarbons with carbon number lower than 6 would not be considered as SOA precursors. Therefore in this revision, we provide supplementary data for low-carbon compounds based on the SIFT-MS quantification to estimate the mass percentage of ethane, ethene, propane, propene and acetylene and their ozone forming potentials, which was not included in previous versions. While such quick online quantification by SIFT-MS might not be as precise as the traditional GC-MS, the results could provide a basic estimation of lighter hydrocarbons.*

We have made revisions in manuscript regarding detailed analysis in different sections:

(1) Main text, 2.1.2 VOCs sampling and analysis, Page 6, Line 15- Line 26:

“In order to complement the quantification of lighter hydrocarbon compounds with 2-3 carbons, selected ion flow tube mass spectrometer (SIFT-MS) was applied, which was recognized as a real-time analytical technique for combustion gases and components in breath (Smith and Spanel, 2010). This part of analysis was done without pretreatments immediately after the gaseous samples from auxiliary engines. VOC samples were transferred from canister to Teflon bag using Agilent headspace syringe with standard volume of 10ml and then diluted with nitrogen. The species that SIFT-MS quantified were not totally consistent with GC-MS. According to the PAMS and TO-15 standard gas applied in GC-MS calibration, 51 species were selected and normalized by mass in SIFT-MS data. Among the 51 species, acetylene, ethane, ethene, propane and propene were the most significant low carbon compounds that were not quantified by GC-MS in this study. Mass proportion of these species was calculated according to the quantification results obtained by SIFT-MS analysis and the impact on ozone forming potentials could be evaluated.”

(2) Main text, 3.2 SOA and OFP by VOCs from ship exhausts, Page 9 Line 22-Page 10, Line 2:

“Considering the impact of low carbon hydrocarbons on ozone forming potential, the quantification

results by SIFT-MS were used to evaluate the underestimation caused by the absence of C2-C3 hydrocarbons in GC-MS quantification. A total of 13 auxiliary engine exhaust samples were analyzed by SIFT-MS and the mass fraction of low carbon VOCs as well as their ozone forming potentials were presented as **Fig.4**.

Fig.4 showed that besides sample 1 and sample 3, the total mass fractions of the low-carbon compounds were below 0.05. Given the fact that SIFT-MS was not able to analyze all 93 VOC species as GC-MS did, the actual fractions could be even lower. When using Maximum Incremental Reactivity scale by Carter's study(Carter, 1994), the calculated OFPs of the low-carbon compounds in sample 1 to sample 13 were between 0.02-0.47 g O₃/g VOCs, with an average of 0.14 g O₃/g VOCs. Compared to the average value of 2.63 g O₃/g VOCs calculated based on the source profile by GC-MS without those hydrocarbons with 2-3 carbon atoms, the underestimation was merely around 5%."

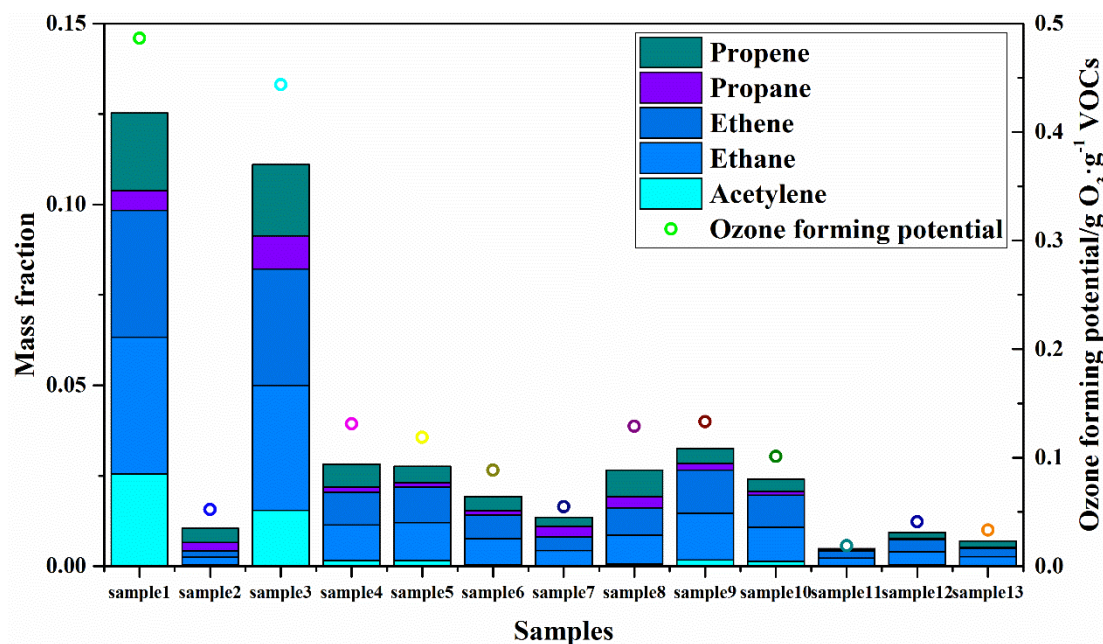


Figure 4: Low carbon VOC mass fraction and corresponding ozone forming potentials.

2. Second, while wind direction is recognized as the most significant meteorological conditions, the atmospheric layer stable conditions should also play an important role in the opportunity of the plumes reaching the measurement site. These factors may have large impacts on the analytical results and conclusions in this study.

Response: Your suggestion regarding the atmospheric layer stable conditions is highly appreciated.

Several key points that were made related to this issue:

- (1) We need to clarify that the major part of this study, the emission profile, is directly measured from the flue pipe not gas plume. So these results were not influenced by the meteorological conditions. To avoid misunderstanding, we clarified where the plumes were used and where the flue were used again.*
- (2) Our previous source apportionment only distinguished results before and after DECA policy implementation. Based on the first round referee's comments, we added discussions on wind direction and did see significant impacts on the results. In this revision, we further analyzed layer stable condition's impacts on source apportionment. We have tried our best to collect as many as possible indicators to identify these impacts. The detailed results were shown in this revision.*
- (3) If the ambient atmosphere is not polluted, the plume will have significant impacts on the capture of air pollutants by instruments. However, our test sites and the test season was with heavy polluted days. And the port is located in Tangshan, a typical industrial city with hourly averaged PM_{2.5} concentrations during our campaign reaching 147 $\mu\text{g}\cdot\text{m}^{-3}$, and over 85% of the total hours under pollution according to the China air quality standard. The main driving force on ambient pollution is not shipping emissions, but intense local industrial pollution and regional pollution background. In this case, when the atmospheric layer becomes stable, both rapid primary particle accumulation and formation of secondary aerosols occurs concurrently. This phenomenon has been frequently observed in North China and become a persistent unsolved issue in academy (see several previous important papers on this issue, e.g. Science Advances(Cheng et al., 2016), PNAS(Wang et al., 2016), ACP(Zheng et al., 2015a;Zheng et al., 2015b)). In conclusion, it is hard to precisely determine the impacts of stable level of atmospheric layer.*

According to the key points above, several detailed revisions have been made in the manuscript:

(1) Abstract, Page 2, Line 3-Line 6:

“The impact of atmospheric stability was discussed based on PM_{2.5} and primary pollutants CO concentration. With the background of frequent haze episodes and complex mechanisms of particulate accumulation and secondary formation, the impact of atmospheric stability would be weakened on the sulfate contribution by shipping emissions.”

(2) Main Text: Page 17, Line 11:

“A new subtitle has been added as **The impact of atmospheric layer stability**”

(3) Main Text: Page 17, Line 12-Page 18, Line 30:

“Apart from wind direction, atmospheric layer stability is also a factor that can affect the particle diffusion in exhausted plumes. However, owing to the experimental conditions and the obscure sampling location during the measurements, the actual planetary boundary layer (PBL) height and cloud cover data were not available in this study, which could directly reflect the atmospheric stability. However, atmospheric layer stability is able to be identified based on indirect parameters. According to the study on the correlation between heavy haze episodes and synoptic meteorological conditions in Beijing, China (Zheng et al., 2015b), the polluted periods were associated with lower PBL height, which meant the atmospheric layer was more stable compared with the clean periods. Moreover, according to Petäjä's study (Petäjä et al., 2016), high particulate concentration would in turn decrease the height of boundary layer via feedback mechanisms. Therefore, it can be inferred that the variation of $PM_{2.5}$ and wind speed can indirectly reflect the changes of atmospheric stability. Additionally, the temporal variation of CO concentrations can also reflect the local diffusion conditions as well as layer stability (Xu et al., 2011), since it is a unreactive air pollutant with a long atmospheric lifetime (two months) (Novelli et al., 1998), and mainly from primary emissions.

Based on the discussions above, temporal profiles of $PM_{2.5}$ and CO concentrations as well as the variation of sulfate contribution from ships are presented in **Fig.11**. The CO and $PM_{2.5}$ data is derived from both online monitoring instrument at the sampling site and national monitoring site 20km away. The trends of data from two different sites show excellent correlation with Pearson's $r=0.91$ and 0.88 respectively for $PM_{2.5}$ and CO (**Fig.11 (a) and (b)**), and this indicates that our measurement results is representative for the ambient atmospheric environment. It can also be observed that the hourly variation of $PM_{2.5}$ concentrations at the sampling site is well agreed with that of CO ($r=0.76$), which illustrates that the local air pollution levels was mainly determined by the meteorology conditions, and the trends of the polluted level near the sampling site can be applied in indirectly judging the stable condition of ambient atmosphere. According to the $PM_{2.5}$ concentration threshold suggested by (Zheng et al., 2015b), hourly air pollution levels during the whole measurement period were classified into four categories: clean (hourly $PM_{2.5} < 35 \mu g \cdot m^{-3}$); slightly polluted ($35-115 \mu g \cdot m^{-3}$); polluted ($115-350 \mu g \cdot m^{-3}$) and heavily polluted ($> 350 \mu g \cdot m^{-3}$). It was found that the hourly averaged $PM_{2.5}$ concentrations during our campaign reached $147 \mu g \cdot m^{-3}$, and over 85% of the total hours were under pollution. Therefore, the atmospheric

layer condition remained stable during most of the time in measurement periods.

While the contribution of sulfate from ships showed an obvious converse trend towards the pollution levels ($r=-0.64$) (**Fig.11 (c)**). The average contributions were calculated as 32.9%, 27.6%, 19.7% and 15.1% during the clean, slightly polluted, polluted and heavily polluted periods, respectively. That is, with the atmospheric stability went up, the contribution of ships would go down.

The explanation to this phenomenon could be summarized in two aspects. Firstly, during the winter haze periods in China, not only the stable atmospheric condition could increase the total concentration of $PM_{2.5}$, but also the secondary inorganic aerosols, especially sulfates, which were rapidly formed via complicated heterogeneous reactions under high relative humidity (RH)(Wang et al., 2016;Cheng et al., 2016;Zheng et al., 2015a). In our study, the RH in port areas were higher than that in the inland, resulting in the predominance of sulfate formation during the polluted period. The effects of the increased ambient sulfate concentrations on reducing the sulfate relative contributions from ships are much more significant than the increased shipping contributions due to stable atmosphere. Moreover, in addition to the local diffusion conditions, the contribution of shipping emissions to the ambient air quality also highly depended on the real-time density of ships at berth. The number of ships at berth would decrease sharply when heavy haze led to fairly poor visibility. Therefore, less shipping emissions lowered its source contributions during the air pollution period. In summary, the effects of atmospheric layer stability on the sulfate contributions from ships was less important during the haze period compared with that in clean days.”

(4) Conclusion: Page 20, Line 14- Line 17:

“The impact of atmospheric layer stability reflected by concentration of $PM_{2.5}$ and primary pollutant CO was also discussed and the results showed that with the background of frequent haze episodes and complex mechanisms of particulate accumulation and secondary formation, the impact of atmospheric stability would be weaken on the sulfate contribution by shipping emissions.”

(5) Figure 11 has been added and the numbers of figures have been revised accordingly.

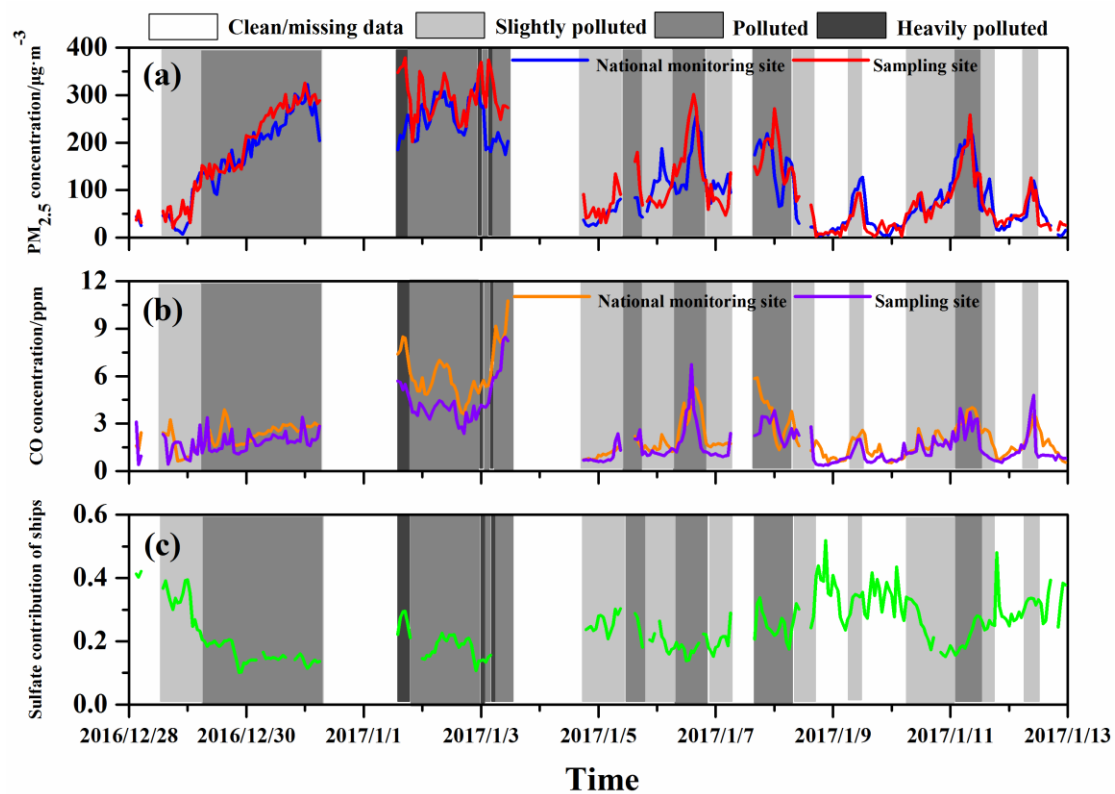


Figure 11: Temporal profiles of PM_{2.5} concentration, CO concentration and sulfate contribution by shipping emissions. (a) PM_{2.5} concentration from two monitoring sites; (b) CO concentration from two monitoring sites; (c) sulfate contribution by shipping emissions.

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