Response to Referee's Comments on acpd-2018-737: " Ambient measurement of shipping emissions in Shanghai port areas"

The authors are pleased to submit our responses to the comments raised by the anonymous referee #2. The authors appreciate and are thankful to the referee's valuable comments and suggestions which help greatly to improve the quality of the manuscript. Each item of the raised comments is responded individually in following pages in the format of:
Referee's comments – Black;
The Authors' responses – Blue.

Response to Referee #2

General comment: The paper regards an analysis of the impact of shipping to atmospheric pollutants measured in the area of Shanghai harbour (China). The approach used is based on the identification and characterization of ship plumes using high temporal resolution measurements of gaseous pollutants and of particles using a SPAMS. The work is interesting and allowed to investigate the typical spectra of particles released by ships as well as to evaluate statistically the contribution of shipping to local air quality. The work is suitable for the Journal and generally well written (even if minor spell check is required), however, some aspects are not completely clear (see my specific comments) and an additional effort in the discussion of size distributions of the impacts should be included. In conclusion, I believe that the paper should be considered for publication after a major revision.

Specific Comments：

1. Title. I think that it is not correct to speak of "measurements of shipping emissions" because emission factors or measurements of specific emission rates are not given. I would suggest to change the title to put in evidence the core of the work: contribution of shipping to atmospheric pollution.

Authors' Response:

Thanks for comment. The authors agree with the referee's opinion about the title of the paper. To reflect the core of this work more accurately, the title of this manuscript would be changed to "Atmospheric pollution from shipping and their contributions to air quality degradation in a port site in Shanghai".

2. Introduction. The adoption of a DECA (Domestic ECA) is quite interesting and it would be even more interesting if a more detailed discussion is included. For example, it would possible to comment on the efficacy of this measure in reducing the impact of shipping on local pollution levels. It is also worth to mention that a recent work (Contini et al., 2015 – Atmospheric Environment 102, 183-190) showed that application of "domestic" restrictions on the fuel quality could be effective in reducing not only local SO_2 concentrations but also primary emissions of particles from ships. I believe that a discussion on this aspect would be appreciated by the readers.

Authors' Response:

We acknowledge the referee's suggestion to include a further discussion on DECA. In fact, the DECA strategy has not only been implemented in YRD region, but also in Pearl River Delta (PRD) and Bohai-Rim area, which constitute the three major shipping regions in China. According to

monitored data at several sites adjacent to port area in YRD, there has been >20% reduction in ambient SO_2 concentration in the same period before and after the DECA measure, although part of the SO_2 reduction are attributable to emission control measures in coal burning in power plant, boilers, furnaces and domestic use in China. There is a published study which dealt with the effectiveness of DECA in PRD region, estimating that the DECA measure could result an average reduction of 9.54% in SO_2 and 2.7% in $PM_{2.5}$ in land areas (Liu et al., 2018).
In the introduction of the manuscript a discussion of this issue will be added as follows:

"…which has taken effect on April 1, 2016. This limitation level of sulphur is still higher than the implemented legislation in many harbors/ports in Europe and US (0.1%) (IMO, 2017). The DECA measure was currently implemented mainly in three major shipping areas including PRD, Pearl River Delta - PRD, and Bohai Rim region in China. Efficiency of the ECA measures has been tested in other places (Contini et al., 2015;Merico et al., 2017). It was shown that the control strategies in sulphur in fuel could generate synergetic reduction in both SO_2 and primary PM release from ships. The benefits of DECA measure in YRD were also suggested by SO_2 concentration at several monitoring sites near the port (>20% reduction after the measure). There is a published study which dealt with the effectiveness of DECA in PRD region, estimating that the DECA measure could result average reduction of 9.54% SO_2 and 2.7% $PM_{2.5}$ in land areas(Liu et al., 2018)."

3. Sections 2.3 and 2.4. It is often mentioned the high temporal resolution of SPAMS measurements, I would suggest to explicitly report the numerical value.

Authors' Response:

SPAMS is a real time measurement and particles drawn into instrument and analyzed consecutively. Depending on the analysis objective, the temporal resolution of SPAMS can be set to several minutes and this value is adjustable.

A notification of SPAMS temporal resolution is added in section 2.3 'Single particle aerosol mass spectrometer (SPAMS)' as:

"The temporal resolution of SPAMS is on order of minutes to accumulate significant number of particles, and this value is adjustable in post data analysis".

4. Page 6 (lines 1-5). V-particles measured without the presence of SO_2 peaks are interpreted as due to the use of low-sulphur content fuel, however, it would not be possible that they are coming from other industrial (or anthropic in general) sources? Some words on this should be included.

Authors' Response:

Thanks for the valuable suggestion. The authors have not considered the fact that vanadium could also be released from other sources as petroleum refinery and other industries. There is indeed petroleum company in adjacent regions and other industries, whose influences should be considered. According to the measurement data, however, the occurrence probability of vanadium particles plume without SO_2 peak is certainly small (3% in cases), so that the interferences of industrial sources will not greatly affect the results in this paper. In another aspect, the determination of contribution of shipping emissions will exclude the industrial influences by confining the wind directions only from the port sector, so that the industrial interferences (from land directions) were set to a minimum.

The original manuscript will be revised as:

"…are indeed mount up quickly. The occurrence probability of this kind of event is low (3% in cases).

The causes of this kind of events are two-fold: firstly, it is maybe due to the anchored ships burning low Sulfur content oil (<0.5 % m/m) to comply with regulations in the port region, which came into force on April 1, 2016; secondly, it is also possible that the vanadium particles be emitted from industry sources such as petroleum refinery companies in this region. The wind directions when these events happened support both of the proposed causes.".

5. Page 7, line 23. To speak at this level of BC is not really useful, likely authors mean EC.

Authors' Response:

Thanks for the penetrating comments. 'BC' is replaced by 'EC' in manuscript.

6. Page 8, lines 2-3. This sentence is not clear and should be re-written. I believe that authors means that ultrafine particle concentrations could be a better metric compared to mass concentrations to investigate the impact of shipping to atmospheric aerosol.

Authors' Response:

The original sentences were revised as:

"…Since the size and mass of fresh exhaust particles are small, the mass concentration PM from exhaust pipes would be inappropriate to represent their real mass contribution after atmospheric aging. This study suggests, as other authors did, that particle number concentration (PNC) be adopted to fully characterize primary ship emitted particles."

7. Page 9, line 30. The approach based on this formula was originally developed in Contini et al (Journal of Environmental Management 92 (2011) 2119-2129) and successively used by other authors. I believe that it would be fair to mention this aspect.

Authors' Response:

This item of reference was replaced by the suggested one.

The original sentence was revised as:

"The calculation method of ship contributions is based on the extraction of ship emission plumes from background concentrations of pollutants, which was originally developed by (Contini et al., 2011):".

8. Looking at the size distributions reported in figures 4 and 6, it appears that V particles are especially relevant for ultrafine particles, however this aspect is not deeply investigated on the evaluation of the impacts. It would be possible to use the approach discussed on page 9 to investigate the size dependency of the impacts of shipping, eventually estimating the impacts for different size ranges. I believe that, if a sufficient statistics could be obtained, this will give very useful additional information compared to the impact on total particle number reported in Table 2.

Authors' Response:

Thanks for that suggestion. The authors agree with the advice to separate V particles into different size ranges and evaluate their impact individually. After inspection of V particle size distribution, the particle diameters will be grouped into three size ranges: <0.4μm;0.4-0.8μm;>0.8μm; Their impacts will be calculated and discussed as a function of size. The next version of manuscript, which will be submitted soon, will cover this topic together with discussions as suggested in 9# comment raised by the referee.

9. Page 10, lines 13-23. The comparison with shipping impact measured in other ports is certainly interesting, however, it is done on relative impacts and not on absolute contributions due to shipping activities this means that it depends not only on ship traffic but also on the contributions of the other sources acting on the specific measurement site. This should be

mentioned because it could explain some of the apparent discrepancy mentioned by the authors. In addition, I would suggest to expand the comparison to other ports analysed with the high temporal resolution approach (Merico et al Transportation Research Part D 50 (2017) 431–445) but also with other complementary approaches (see for example Viana et al 2014 Atmos. Environ. 90, 96–105).

Authors' Response:

The authors agree with that advices. We acknowledge that the pollution absolute contributions from shipping are also important. The discussions with other port will also include more relevant studies in the literatures. Absolute contributions of shipping emissions and relevant discussions will be embodied in the next version of manuscript which will soon be submitted.

10. Regarding the impacts reported in Table 2, it would be possible to estimate the uncertainties?

Authors' Response:

In the preparation of Table 2 we have considered the estimation of uncertainties, which is a conventional practice in scientific report. The uncertainties of calculation may stem from sources such as the identification of plumes, the definition of port sector directions and the gaseous and particulate measurement itself. Some of these sources are found difficult to define. To be consistent with the original study (Contini et al., 2011), the uncertainties in this work will be estimated by inspection variations in slight changes of wind direction and the elimination of data of low wind velocity (< 0.5 m/s). The uncertainties will be included in the next version of manuscript.

11. Page 11 line 5. This sentence is not clear. Authors likely mean that the impact of shipping is more relevant and clearly discernible on SO_2 and V particles compared to the other pollutant analysed. Could authors clarify?

Authors' Response:

Thanks for the advice. The original sentence would be revised as:

"The concentration of SO_2 and vanadium particles has displayed better relevance to shipping emission in the coastal port than other pollutants analyzed, such as NO_x and $PM_{2.5}$ concentrations".

12. In the supplementary material it is reported "…in present study the online single particle measurement was utilized to indicate the occurrence of shipping emission Plumes…" however in the main text was mentioned that both particles and SO2 concentrations were used. Please clarify this apparent contradiction.

Authors' Response:

Thanks for pointing out this unclarity. The intention behind the supplementary text is to give the reader a extended discussion on the identification method of ship emitted particles. As explained in supplementary text, the adoption of vanadium tracer could not guarantee that every single ship emitted particles in a plume be identified. From figure S1 it could be inferred that only a fraction of ship emitted particles in plumes are identified by vanadium peak criteria, because not every individual particles in a emission plume contain a detectable vanadium content.

The mentioned sentence "…in present study the online single particle measurement was utilized to indicate the occurrence of shipping emission plumes…" is emphasizing this fact and not really mean that only the vanadium particles were used to indicate the influence of plumes. Actually both of vanadium particles and SO_2 concentration were applied to identify plumes in data analysis process. In fact, the SO_2 concentration is critical to identify plumes in which few vanadium particles

were present.

To prevent possible confusion, the original sentence is revised as :

" … in present study the online single particle measurement, together with synchronous SO_2 concentration, was utilized to indicate the occurrence of shipping emission plumes…".

Minor corrections

13. Page 1, line 15. Better "particle size distributions".

Authors' Response:

Suggestion accepted.

14. Page 1, line 28. Please eliminate the initial S.

Authors' Response:

Accepted.

15. Page 2, line 19. Subscript for SO_2. The same in page 4 (line 25).

Authors' Response:

It has been corrected.

16. Page 7, line 14. Better "different size distributions…".

Authors' Response:

Accepted.

17. Page 8, line 12. Better "by the dominant". In addition, I would remove etc, if necessary please mention explicitly.

Authors' Response:

Suggestion accepted. The sentence is revised as:

"In the positive mass spectra the V-OC type are characterized by the dominant organic peaks like $C_2H_3^+$, $C_2H_5^+$, $C_2H_3O^+$,…".

18. Page 8, line please remove etc. as above.

Authors' Response:

The 'etc.' is removed.

19. Page 8, line 25. Better "is therefore not attempted…"

Authors' Response:

Accepted.

20. Page 9, line 2. > 0.5 μm

Authors' Response:

Original letter replaced with the Latin letter 'μ'.

21. Page 11, line 10 ozone without capital letter.

Authors' Response:

Accepted.

Refereces

Contini, D., Gambaro, A., Belosi, F., De Pieri, S., Cairns, W. R. L., Donateo, A., Zanotto, E., and Citron, M.: The direct influence of ship traffic on atmospheric PM2.5, PM10 and PAH in Venice, Journal of Environmental Management, 92, 2119-2129, 10.1016/j.jenvman.2011.01.016, 2011.

Contini, D., Gambaro, A., Donateo, A., Cescon, P., Cesari, D., Merico, E., Belosi, F., and Citron,

M.: Inter-annual trend of the primary contribution of ship emissions to PM2.5 concentrations in Venice (Italy): Efficiency of emissions mitigation strategies, Atmospheric Environment, 102, 183-190, 10.1016/j.atmosenv.2014.11.065, 2015.
IMO: Emission Control Areas (ECAs) designated under MARPOL Annex VI, 2017.
Liu, H., Jin, X., Wu, L., Wang, X., Fu, M., Lv, Z., Morawska, L., Huang, F., and He, K.: The impact of marine shipping and its DECA control on air quality in the Pearl River Delta, China, Science of The Total Environment, 625, 1476-1485, https://doi.org/10.1016/j.scitotenv.2018.01.033, 2018.
Merico, E., Gambaro, A., Argiriou, A., Alebic-Juretic, A., Barbaro, E., Cesari, D., Chasapidis, L., Dimopoulos, S., Dinoi, A., Donateo, A., Giannaros, C., Gregoris, E., Karagiannidis, A., Konstandopoulos, A. G., Ivosevic, T., Liora, N., Melas, D., Mifka, B., Orlic, I., Poupkou, A., Sarovic, K., Tsakis, A., Giua, R., Pastore, T., Nocioni, A., and Contini, D.: Atmospheric impact of ship traffic in four Adriatic-Ionian port-cities: Comparison and harmonization of different approaches, Transportation Research Part D-Transport and Environment, 50, 431-445, 10.1016/j.trd.2016.11.016, 2017.