EDITOR'S REVIEW OF MANUSCRIPT ACP-2018-737 – REFERRING TO MANUSCRIPT VERSION 03:

RESPONSE TO COMMENTS FROM REFEREE #2

The authors have sufficiently responded to the comments of Referee #2. The referee has accepted their responses without requesting further modifications.

RESPONSE TO COMMENTS FROM REFEREE #1:

To start with, I repeat my email from 19 November 2018, in which I responded to the authors with respect to their inadequate response to the comments raised by Referee #1. In that email I wrote:

"As the handling co-editor of your manuscript acp-2018-737, I am investigating the current situation of this manuscript after the public discussion phase has been closed and you have responded to the referee #2, resulting in an immediate response by referee #2 that he/she will reject your manuscript in case you do not respond in an appropriate manner. Actually the manuscript is stuck.

I analysed the review provided by referee #1 and found it detailed and appropriate, and written in the usual way, good reviews are provided. In particular I cannot see any unfriendly, insulting remarks.

If you wish to move on towards the successful publication of your manuscript in ACP, I urgently recommend that you respond to the reviewer's remarks in a more open way. In particular, referee #1 has requested better explanation and discussions of results in some places and your response was, e.g., "will be presented and discussed in next edition". However, such a response is insufficient since it does not allow the reviewer to assess whether his or her request has been answered sufficiently."

In the light of this request for clear response, I analysed the authors' responses to the comments by Referee #1, as submitted on 4 January 2019. Most of the minor comments by Referee #1 have been answered appropriately. However, major and more fundamental concerns have not been answered at all or in an inadequate manner. Therefore I request the following changes before the manuscript can be accepted.

Before going into details I want to remind the authors that referees and co-editors work on a voluntary basis. They spend extra time to help reviewing publications and ensure journal quality standards. The authors' responses to referees' comments and suggestions should always reflect the additional work load the referees and co-editors have accepted!

SPECIFIC CONCERNS RAISED BY REFEREE #1

Detailed points are listed below. For clarity I repeat from the authors' responses to the referees as follows: Referee #1 comments (in black), authors' responses (in blue), and changes to the manuscript (in red). Editor's remarks are added in red italic letters.

REFEREE #1: It is not clear how the components in particles such as vanadium were identified and quantified by the SPAMS. Detailed information is needed.
AUTHORS' RESPONSE: SPAMS identify particle composition, such as vanadium, in mass spectrometry method. In the ionization laser beam in SPAMS, the components in particles are ionized into ions carrying charge, then they are separated in the Time-of-Flight tube by atomic mass of the ion. Lighter ions, such as H+, transit fastest in TOF tube and produce peaks in shortest time. The 208Pb+ is heavier so that it reach the MS detector with longer time. This will result a spectrum sorted by ion atomic mass. Components of different atomic mass produce peaks in different position in mass spectrum. Vanadium in particles normally produce peaks at mass = 51V+ and 67 (VO+) and thus can be identified. This is the basics of MS and unnecessary to be included.
CHANGES IN MANUSCRIPT: None

EDITOR'S REMARK: The authors explain basic explanation of how a MS works, which indeed can be expected as basic knowledge. However, Referee #1 requests detailed information about the quantification of Vanadium concentrations. This needs to be added.

REFEREE #1: Page 5, lines 21-25: The discussion here is questionable. By looking at Figure 2, whenever ship plumes were captured, both NO and SO2/vanadium levels were high and correlated well. On what basis, the authors claimed the NOx in plumes reaching the site was aged? Using the NO/NO2 ratio in the plumes? Compared to the ratio measured in other countries and probably different type of ships? This is not convincing. Besides, NO2 is also emitted from shipping as a primary pollutant.
AUTHORS' RESPONSE: We have not tested NO/NO2 ratio in the exhaust because the observation was carried out in a station on land. The author also know that NO2, together with NO, is released as primary emissions. However their ratio, upon their emission into atmosphere, will subject to change quickly through the oxidation of NO into NO2 in the existence of ozone (O3), which is abundant in summer time. This was evidenced by the quickly reduced O3 level during plumes in Fig. 2 in manuscript. The referee seems to be doubtful about this reaction, which is the very basis that the NO-NO2-NOx analyzer is working on.
CHANGES IN MANUSCRIPT: *None*

EDITOR'S REMARK: I fully agree with Referee #1 who requests are more careful discussion. The measured levels of NO_x and the separation into NO and NO_2 cannot be used to identify ageing of ship plumes. Since the authors do not know the level of chemical processing (depending on O_3, meteorological conditions and atmospheric radiation) of ship exhaust in the marine boundary layer during transport to the sampling site, it seems to be more appropriate looking only at the sum parameter NO_x. This suggestion is supported by the results presented in Table 1. Looking at NO there is no statistically significant difference between in-plume, non-plume and port average cases. An almost similar statement holds for NO_2.

REFEREE #1: Given that shipping emission is a major source of PM2.5, it is odd that no PM2.5 peaks were found during the ship plumes in Figure 2. The reason provided by the authors is quite confused. Is it because ship emits sub-micron particles or because the malfunction of the PM2.5 monitor?
AUTHORS' RESPONSE: The authors have stated that PM2.5 peaks was not as apparent as that of SO2 and NOx in plumes, and have not states that no PM2.5 peaks were found. It should be made clear that only a short period of data was shown in Fig.2 only with the purpose of illustrating temporal variation in plumes. In the last question the referee is suspecting that PM2.5 analyzer was in malfunction. The author would like to illustrate another longer period of data in Figure R2 (shown below) and let the referee make his judgement. The sharp peaks of SO2 in Figure R2 could help to locate the plumes. If the PM2.5 instrument is in malfunction, how did the PM2.5 instrument happened to malfunction only in plumes? In another aspect, the SPAMS particle number

concentration have shown good correlation with the PM2.5 measurement. How did the two instruments both malfunction? It sounds ridiculous.
CHANGES IN MANUSCRIPT: *None*

EDITOR'S REMARK: The authors' response to the referee is not acceptable. Moreover, the entire discussion of the $PM_{2.5}$ mass concentrations is confusing and inconsistent. Referee #1 simply requests are more consistent presentation of the $PM_{2.5}$ case, which I do as well. We know from earlier observations that most of the PM emitted by ship engines is far smaller than 1.0 µm in diameter and will thus not contribute significantly to the $PM_{2.5}$ mass concentration. This fact needs to be pointed out very clearly. In that respect the results presented in Table 1 are consistent but the explanation in the text is confusing and requires careful revision. In particular, the explanation announced on Page 7 line 21 of the annotated manuscript is either missing or well hidden.

REFEREE #1: The reason for absent SO2 is contradictory to Figure 2. If the ships complied with the new regulations, why would you still see SO2 peaks in ship plumes? This kind of discussion is misleading.
AUTHORS' RESPONSE: As stated above, the cases the SO2 peaks were absent were rare and will not affect significantly the result of the study. The new regulation only confine the Sulfur content in fuel, not eliminate the Sulfur from the fuel.
CHANGES IN MANUSCRIPT: *None*

EDITOR'S REMARK: I fully agree with Referee #1. If the authors do (for 3% of the cases) not find a correlation between SO_2 and V, how have these cases been treated? Where they excluded from the analysis? Clarification is requested.

REFEREE #1: It does not make sense to compare a site near sources with sites in a city without any detailed characteristics of the locations. It would be more meaningful to compare the ship emissions in this study with other similar studies conducted in Shanghai. In fact, there are a number of ship emission studies in this city.
AUTHORS' RESPONSE: The comment is not acceptable. We made comparison between air pollution level in a port site with that at the urban area of the same city, How did it make non sense? The referee had better suggest a study which he think make more sense.
CHANGES IN MANUSCRIPT: *None*

EDITOR'S REMARK: This response it not acceptable. It is not the duty of the referees to make the authors familiar with current literature! I fully support Referee #1 request for a comparison of the presented results with those from other studies in Shanghai port.

REFEREE #1: the explanation of low PM2.5 levels at the port site is not convincing at all. Why would other pollutants from shipping emissions be higher if this was caused by clean air?
AUTHORS' RESPONSE: The slight lower PM2.5 concentration at the port site is a fact. The author only postulated possible explanations.
CHANGES IN MANUSCRIPT: *None*

EDITOR'S REMARK: The authors have to provide consistent explanations for their observations. There is no reason why $PM_{2.5}$ should be influenced by clean air why gaseous pollutants will not. Another explanation is needed here.
Another remarks concerns Figure 6, which also shows the inconsistency of presentation of PM results. In the figure caption, the authors indicate that particle number concentrations are presented. In the figure itself, however, the y-axis title (left panel) indicates "Particles/hour". This is definitely not an

adequate unit for particle number concentrations! Furthermore, the y-axis of the upper right panel indicates "Particle number", whereas the authors mean particle number concentration in units of cm^{-3}*. Both issues need to be clarified.*

MINOR ISSUES:

- *The company's name providing the instruments is THERMO SCIENTIFIC, not THERMAL SCIENTIFIC.*
- *Furthermore, another language editing is mandatory before publication.*