

Review of “*Influence of vessel characteristics and atmospheric processes on the gas and particle phase of ship emission plumes: In-situ measurements in the Mediterranean Sea and around the Arabian Peninsula*” by Celik et al, submitted to ACPD.

In their manuscript, the authors present data and analysis of measurements of over 250 ship emission plumes in the Mediterranean and around the Arabian Peninsula. From the data, the authors present information on the dispersion of the ship plumes, as well as comprehensive data on different components of the emission plumes. The manuscript also connects these observations to ship characteristics obtained from the AIS system. Finally, the authors also discuss the different atmospheric processes that affect the various components of the plume.

The manuscript's topic is very well suited to Atmospheric Chemistry and Physics, and the data and its analysis are interesting and of potentially high value. The paper is well structured and easy to read. Therefore, in principle the paper should be considered for publication in ACP. However, I think there are a few mistakes in the methodology and the reasoning behind drawing some conclusions, especially with a) the interpretation of the black carbon (BC) data and b) the computation of the O:C ratios. These are significant enough that they must be corrected before the paper is published in ACP, as they might change some of the presented results.

In the following, I will first discuss these two possible issues, and then give other general comments on the manuscript:

1. BC data dependence on pressure and temperature

I have serious reservations regarding the conclusion driven from the BC data dependence on ambient temperature and pressure (if I understood correctly). To my understanding, the ambient temperature and pressure only affect the gas concentration (in units of molecules/m³) and don't affect the fraction of oxygen in a given mass of air. As in a diesel engine the air is in any case highly compressed, the amount of oxygen available should not change too much. There certainly is the possibility that there might be other pressure/temperature effects that affect engine operation, but to use p/T as a proxy for these, the effects should be more thoroughly explained and appropriate references given.

Additionally, I see a potential alternative explanation to the observation: I'm not fully aware on how the flow calibration for the BC instrument has been performed. However, If the instrument pump pushes a constant mass (or constant number of molecules) per unit time through the filter, i.e. $n = pV/kT = \text{constant}$ (as would be the case with a mass flow controller), then as the value of p/T increases, the volume going through the filter decreases. As the derived BC concentration depends in a non-linear way (as explained in Drinovec et al., 2015, which the authors also cite) from the change in attenuation and also the volume flow rate, I think it is conceivable that the observation in Fig 3 is caused by this effect. This should be considered seriously, especially with the reported periodicity of the instrument reading with respect to the measurement container temperature control.

I would suggest reviewing and re-analysing the BC data and its conclusions. For example, it would be informative to see if the BC concentration vs. p/T -dependence is visible only for plumes, or also for ambient BC concentration measurements.

2. Calculation of O:C ratios

I had some trouble following the definitions in eqs 3-5; I think there's an error in the definition of the terms. Assuming that O/C and H/C ratios refer to the atomic ratios, the second version for the computation of the concentration of [O] and [C] are wrong. This can be illustrated with an example: an organic compound with 5 oxygens, 3 hydrogens, and 3 carbons, so we get the following values

Compound	N	M _x	m _x =M _x •N
O	5	16	80
H	3	1	3
C	3	12	36

The total mass (i.e. [organic]) is 119, and O/C = 5:3 and H:C = 3:3.

From this example, we would get

$$[O] = [\text{organic}] \frac{m_O}{m_O + m_H + m_C} = [119] \frac{80}{80 + 3 + 36} = [119] \frac{80}{119} = 80.$$

According to eq. (3), this should be equal to

$$[\text{organic}] \frac{\frac{O}{C} \cdot M_O}{\frac{O}{C} \cdot M_O + \frac{H}{C} \cdot M_H + \frac{C}{C} \cdot M_C} = [119] \frac{\frac{5}{3} \cdot 80}{\frac{5}{3} \cdot 80 + \frac{3}{3} \cdot 3 + \frac{3}{3} \cdot 36} = [119] \frac{5 \cdot 80}{5 \cdot 80 + 3 + 36}$$

$$= [119] \frac{400}{439} = 108.43$$

which gives an error of 35% compared to the first equation for [O].

For these calculations, it is also unclear, which of the equations was used for the actual computation of the values. This should be clarified, and in case it has an effect on the results, the new results presented in the revised manuscript.

3. General Comments

- Pg.2, line 1: "The results enable identification of...": I think the identification has already been done, so this should be reflected here.
- Pg 4, line 14: "...appropriate inlet systems": The sampling lines can have profound effects on the measured quantities, so I think it would be good to elaborate a little on the sampling line system. If some known guidelines for the sampling were followed, a reference could be given. Are the losses given in Table 1 the line losses? If yes, were they calculated or experimentally determined?
- Page 5, line 15: I think it could be good to clarify the PM1 calculation algorithm: I think based on the explanation it is

$$PM1 = V_{FMPS} * \rho_{AMS+BC} * F_{corr}$$

where V_{FMPS} is the volume obtained from FMPS, $\rho_{\text{AMS+BC}}$ is the average density obtained from AMS and BC measurements, and F_{corr} is a correction factor that mostly accounts for underestimated concentrations >130 nm. As the correction factors are averages estimated from the data, I think it would be good to give some information on the variation, eg. give the standard deviation of $\rho_{\text{AMS+BC}}$ and F_{corr} . Also, if the OPC was measuring large particles, should it be included in Table 1?

- Page 5, line 31: “..linear or Gaussian fits.” Here it is unclear what was fitted to what. Could this be elaborated?
- Page 8, line 11: “..linearly interpolated”: I think this is just taking the mean background from before and after the plume. Is this so?
- Line 14-15: “defined as ...”: it would be easier to read this just as a formula I think.
- Page 9, line 4: “dispersion time constant”: As there are several ways of defining time constants, I think it is important to say which one is meant. Is it the concentration half-life, or e-folding time, or some other one?
- Page 9, line 13: “one standard deviation”: I think this should be geometric standard deviation
- Page 9, line 20: “measured OH concentrations were not used... only describe the situation and the research vessel’s position and not within the plume”. Is this not also the case for the photolysis rate?
- Page 12, line 5-10: A reduction of particle number EF was observed with vessel size, and this is attributed to coagulation. Is this the only explanation, or could there be others too? What is the variation in the residence time?
- Page 12, line 12, and pg 19, line 7. -> I find the finding on the wind speed influencing the effective EF interesting. I think this is an important effect to take into consideration. However, I think that it should be made clear that at the source, the emission is the same – the difference in the observed EF is introduced only later during the processing of the plume. Additionally, I’d like to know the more detailed reasoning behind this, as to me, the increased dilution in at high WS should be accounted for in the calculation. Is the age of the plume (since emission) factored in here? Does the wind speed affect engine operation?
- With regard to the wind speed, was there a correlation with the plume dispersion time constant (page 9) and wind speed? As the explanation for the effect on particles is related to dilution, I think there might be a connection between the two. If not, why?
- More generally, I think that the term emission factor (EF) should be used only for actual emission at the point and time of emission, and if the observed aerosol after some time has some differing characteristics, this should be noted with some qualifier, e.g. ‘apparent’ or ‘effective’.
- Figure 2: I have some trouble understanding the decision-making when two ships are in the path of an air mass. In the example given, it seems to me from the figure that both the red and the blue ship are relevant for the observed plume. Why was the blue ship not considered? This could be clarified.