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Comment

## ***Interactive comment on “Physical and chemical characterisation of PM emissions from two ships operating in European Emission Control Areas” by J. Moldanová et al.***

**Anonymous Referee #2**

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Moldanova et al. present results of measurements of particulate emissions (and some gas emissions) from two ships, with samples from the main engine (ME) and auxiliary engine (AE). For one ship, measurements on the ME were made at two different engine loads using heavy fuel oil (a high sulfur fuel). For the second ship, they made measurements at one engine load for two different fuels, HFO and marine gas oil (a lower sulfur fuel). Measurements such as these are important, given the impacts of PM emissions from ships on air quality and climate. Overall, the measurements are comprehensive in their scope, with emissions of many different PM contributors being examined. Given the variety of emitted species considered and the various ship oper-

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ating conditions, I found it to be, at times, a bit confusing which particular ship/condition was being discussed, or what was being compared with what. I suggest that the authors aim to make their discussion a bit more focused and clear. I have a number of concerns, provided below, regarding such issues of clarity. And I have some scientific concerns regarding appropriate treatment/consideration of measurement uncertainty. I believe that ultimately this paper will be publishable, but that substantial modifications are needed at this point.

Specific Comments (in mostly chronological order):

I find the abstract to be quite lengthy and wordy (especially the first paragraph). I suggest shortening, as well as re-writing for grammar.

Abstract: The uncertainties reported for EF(OC) for MGO are too small and do not account for experimental uncertainty.

Abstract: The authors should use consistent units. For example, EF(OC) is given in g/kg-fuel while EF(SO<sub>4</sub>) is given in mg/kg-fuel, despite the similar magnitude.

P3939/L2: The authors should clarify that the EEPs measures particle mobility diameter (there are many different types of particle diameter than can be measured and this becomes particularly important for particles that contain substantial black carbon fractions and may not be spherical). Similarly, the authors should clarify that the GRIMM measures optical diameters (which again may be particularly problematic for very “black” particles, depending on the way in which the instrument was calibrated. The diameters measured by the EEPs and by the GRIMM are likely not equivalent.

Section 2.3: Is the uncertainty for EC the same as for OC (31%)? Typically they are not. How was this 31% uncertainty arrived at? The authors used transmission mode corrections of pyrolysis artifacts. This can give different results than reflectance. This should be (very briefly) discussed in terms of how it contributes to their uncertainty estimates?

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Section 2.3: The authors need to provide a reference to the US EPA document that suggests the use of a mass absorption coefficient of 12.6. Further, units should be provided (e.g. m<sup>2</sup>/g)

Section 2.3: I find the discussion of the DTT assay method to be quite unclear, and I do not understand the justification for the use of the total PM mass sampled on the quartz filter given that only the extractable portion was analyzed. Specifically, some of my confusion arises because on the top of p3943 the authors mention EC as an important aspect of DTT activity, but the extraction and filtering process very likely biases the results away from EC.

P3946/L29: The authors conclude that EF(CO<sub>2</sub>) does not exhibit any dependence on engine load. However, both the EF(CO<sub>2</sub>) and EF(SO<sub>2</sub>) very clearly decreases (from 678 to 617 g/kWh for CO<sub>2</sub> and from 3.65 to 3.24 for SO<sub>2</sub>), inconsistent with the statement. Granted, there are only two points for each, but if the authors estimates of uncertainty are actually correct then they are statistically different.

Section 3.1: I find it is often difficult to tell when the authors are referring to the main engine or auxiliary engine in their discussion. For example, the discussion on P3947 regarding PM emissions makes it sound (to me at least) like the authors are saying that EFs from the main engine were larger for HFO operation by S1 compared to S2, but similar “for the two investigated engines.” But there are no measurements for the ME on S1 operating on MGO, only for the AE. This is just one example of what I find to be the ambiguity in the discussion. It might be helpful if the authors split their discussion into ME and AE separately.

Fig. 2: Why is TSP » PM<sub>10</sub> in some cases? What does this say about measurement uncertainty?

P3948/L4: The GRIMM and EEPS cannot be compared by simply looking at the overlap range in the size distributions. As mentioned above, these do not measure the same “size” (one being mobility, one being optical) and thus this must be accounted for

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(which is difficult to do).

Figs. S1 and S2: Although the photos are nice as reference, most likely more useful to the general reader would be a schematic diagram showing the flow paths directly.

P3948: The EF(PN) should also be compared with the observations from the various work by Lack et al. (JGR, 2009 and ES&T, 2012).

Fig. 4: The authors comment on some reasons for the difference in the observed size distributions (especially mass-weighted) between the main engine at two loads for S1. However, they provide little/no discussion of why the AE would show similarly such a large contribution from particles with optical diameters > 1000 nm as at the high load in the main engine. I find this behavior somewhat counterintuitive, especially given the arguments advanced for the difference between the high/low load conditions (greater reentrainment of particles from the walls for high load compared to low load). I would, perhaps naively, think that the AE would behave more like the ME at low load and thus show greater similarity. This should be discussed.

Fig. 4: The authors also seem to only discuss (P3949) differences between the high/low load experiments in terms of the behavior of the large particles. But there are also substantial differences in the number of the smallest particles. This should be discussed.

Volatility (P3949): Presumably, volatility is assessed by determining the number concentration of particles that remain after thermodenuding relative to the non-thermodenuded case (this is never explicitly stated). Does this account for losses of particles in the thermodenuder that might occur separate from heating, but simply due to wall losses? In some thermodenuder types, the loss of small particles can be extensive even for non-volatile particles. See for example Huffman et al. (2008, AS&T). The authors should provide some discussion of the accuracy of their measurements regarding particle losses. Have size-dependent transmission factors for non-volatile particles been measured for their thermodenuder type? This, to me, seems critical to

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understanding their reported numbers in the context of the literature. Again, losses of non-volatile particles are likely to be size-dependent, and thus if the size distribution here is different than for the literature studies, and if size-dependent losses are not accounted for, then this could be a simple explanation of the different results.

Section 3.3: The authors state that “relatively large uncertainties” associated with mass closure come about because EC/OC was measured on different filters than sulfate and other species. But this should come out in the actual uncertainty estimates, if (1) uncertainties are being properly determined and (2) if uncertainties are propagated through completely. The authors do not actually even present numbers associated with mass closure to allow the reader to assess how good/bad the closure is. This information should be provided.

P3950/L2: I do not understand the statement “The ash residuals left on quartz filter punches after the temperature program of the Sunset analysis could not be defined due to uncertainties in the gravimetric analysis of these punches.”

Fig. 6/P3950: How was sulphate associated water determined? This is not stated.

Fig. 6/P3950: It is never stated where the “unidentified mass” (UIM) comes from. Is this the difference between the total mass and the sum of individual species? Or some other thing? If the former, how can this be reconciled with the earlier statements regarding the large uncertainties in the mass closure?

P3950/L11: I am not surprised that the thermodenuder estimate of volatiles differs from the gravimetric and chemical based analyses given that the authors do not state whether they account for loss of non-volatile particles in their thermodenuder and because the authors (1) do not know what density to use for their particles when translating the number distributions to mass distributions for the thermodenuder experiments and (2) as the volatile material is removed the particles are likely to become increasingly fractal-like, making the conversion between measured diameter and mass more tenuous because the EEPS measures mobility and the GRIMM optical diameter.

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Uncertainties in EF's: The reported uncertainties MUST be at least as large as the uncertainties associated with the measurements. Here, they seem to be roundly underestimated. Consider as an example the reported EF(OC) for one case was  $0.64 \pm 0.12$ . However, the stated uncertainty in the OC measurement was 31%, which means that the MINIMUM uncertainty is  $0.64 \times 0.31 = 0.20$ , not 0.12. And this only accounts for the uncertainty in the OC measurement, not the ancillary measurements that are necessary to convert the measured OC to an actual EF. I believe that uncertainties for ALL reported EFs must be updated to properly reflect the uncertainty in the measurements.

Fig. 7: Associated with the above comment, I believe that Fig. 7 should be revised to include error bars. This will likely require splitting the OC and EC into separate bars, rather than the stacked bar. Or the authors could propagate the uncertainties for the EC(OC) and EF(EC) and provide error bars on the total.

P3951/L5: What constitutes an “experiment.” For example, multiple PM1 samples for a given engine/fuel/load? Or the PM1/PM10/TSP for a given engine/fuel/load? Further, I don't fully understand the numbers as reported, as there should be a coefficient of variation for each experiment. So are these the average values across all experiments?

P3951/L10: I don't understand why EF(EC) and EF(BC) are not used in place of the “relative contributions”. I would think that the EFs, like the relative contributions, should account for differences in collection times but, more importantly, are the parameters that we are most directly concerned with.

Fig. 8: In my opinion, there is a lot of wasted space on this figure that makes it more difficult to compare between different results. I suggest an upper limit of 40% be used.

P3952: Given much of the discussion on this page, it would seem useful to include a graph of  $EF(X)/Fuel(X)$ , where X is the species of interest. This could be done for each of the species considered where both values (EF and Fuel) are available. Or, this information could be provided in a table.

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P3952: The %conversion numbers given for MGO here seem inconsistent with those reported in the abstract (0.1% vs. 0.01-0.02%, respectively). This should be reconciled.

P3954, L24: I don't see how Fig. 11 provides a "global" view, given that there are only two images. What do the authors mean by "global."

P3954: The authors should define more explicitly what they mean by "primary soot particles" and "agglomerates" so that the reader can know specifically to what they are referring.

Other comments:

I believe that use of sub-headings in section 3.3 would be highly beneficial.

Abstract: Phosphorous (P) is not a metal.

P3935/L18: I find this statement confusing as to what "navigation" means.

P3935/L26: The "only" here is inconsistent with the next sentence.

P3937/L11: RoRo should be defined for a more general audience.

Section 2.1: Funding information belongs in acknowledgements, not in the main text.

P3955, L2: This should actually read Fig. S6. Other supplementary Figure numbers on this page are also incorrect.

It is my suggestion that this work could benefit from some writing assistance from a native English speaker.

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Interactive comment on Atmos. Meas. Tech. Discuss., 6, 3931, 2013.

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