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## **Comment on amt-2020-482**

Anonymous Referee #1

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Referee comment on "Detection of ship plumes from residual fuel operation in emission control areas using single-particle mass spectrometry" by Johannes Passig et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2020-482-RC1>, 2021

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Review to "Detection of Ship Plumes from Residual Fuel Operation in Emission Control Areas using Single-Particle Mass Spectrometry" by Passig et al., AMT-2020-482

This paper reports on the detection of individual ship plumes over distances of several (ten and more) kilometers by single particle mass spectrometry. The authors use laser ablation ionization with resonance-enhanced detection of iron which increases the sensitivity to iron-containing particles. Such iron-containing particles (together with vanadium and nickel) are then used to identify ship plumes. Furthermore, a concentrator was used to increase the concentration of particles.

I have some issues with the scope and the overall goal of the paper. I don't see why it was submitted to AMT. ACP or Atmospheric Environment would have been a better choice, because the measurement technique is not the main focus of the paper. The technique of resonanced-enhanced detection of metals has been described by the authors in an ACP paper (Passig et al., 2020), which to my opinion should have been an AMT paper.

In the present work, neither the resonance-enhanced detection nor the application of the concentrator (which is done in Passig et al, 2020, supplement) are described and discussed. I suppose the authors consider the detection of ship plumes using the enhanced detection efficiency for iron-containing particles as the technical aspect in this work. But I guess the decision whether this manuscript fits into the scope of AMT or not has already been taken by the editor.

I am somewhat impressed by the finding that ship plumes can still be detected by single-particle analysis over such long distances, but the authors have to admit that this was also a good piece of luck and that they are strongly dependent on wind direction. If fact, the authors admit that in the conclusions section.

It has also to be noted that the idea of analyzing ship plume particles by aerosol mass spectrometry is not new. A very similar study by Ault et al (which is referenced here) identified about 12 ship plumes in 5 days. Diesch et al. (2013, not referenced) identified and analyzed 139 ship plumes in 5 days, using a similar setup, but being located much closer to the shipping route. Furthermore, a recent ship-based study (Celik et al., 2020, not referenced) detected more than 250 ship plumes during a 2 month cruise and were therefore able to provide a large data set on ship plume characteristics.

Overall, I am therefore not convinced that this method will be useful to monitor particle emissions from ships on a regular basis. Single-particle mass spectrometers are typically large, heavy, expensive and require a lot of maintenance (please correct me if I am wrong). This together with the difficulty in plume assignment to individual ships will make it very hard to use this setup in a kind of monitoring application.

Nevertheless, the paper presents some interesting data on selected ship plumes. I have two major comments that need to be addressed before publication, and several minor comments and technical corrections as listed below.

### **Major comments:**

1) What I miss the most is a detailed analysis of all detected plumes. In Figures 4 and 5 I count about 12 "enhancements" of the V-Fe-Ni particle class, which (according to the argumentation of the manuscript) are likely ship plumes. In section 3.4, however, only the plumes on June 28 are analyzed further. To be precise, they are assigned to ships but not analyzed. I suggest including (at least) a table with all events that qualify as a potential ship plume, list the possible assignment to a ship and the plume characteristic. Here, ion ratios averaged only over the plume times might be a useful indicator. V/Fe, Fe/Ni, Fe/S, for example. Additional graphs with averaged mass spectra for the plume periods or size dependent composition of the plume particles might also be helpful.

A related question here is: Were there no accompanying data? No CPC, optical particle counter, CO/CO<sub>2</sub>/NO<sub>x</sub> detectors, no black carbon instrument? All such parameters are usually required to better understand the plume characteristics and assignments.

2) It is not clear to me whether the resonance-enhanced iron detection is needed for this analysis. Would you find and identify the the same amount of ship plumes by:

- applying the ART algorithm ignoring m/z 54 and 56, or
- using vanadium and nickel as marker ions without ART clustering? I mean, simply looking at the time series of m/z 51 (V) and m/z 58 and 60 (Ni)?

I think that in an AMT paper that is supposed to be technical this should be shown and discussed.

### **Minor comments**

Lines 56-57: UAVs are also limited in their payload

Line 61: Gas and particle both mix with air and are transported. "in contrast" seems not to fit here.

Lines 107 - 118: This sounds like "normal" SPMS analysis. But the resonance-enhanced Fe detection will lead to an overweighting of Fe. Is this considered in the analysis? Please comment and discuss.

lines 120-122: This should go into section 2.1 (line 98 ff)

Lines 123 - 125: The HYSPLIT trajectory analysis is not sufficient. In the supplement, we see only single trajectories. It is necessary to use the "ensemble" option or to manually initialize more starting times and locations to get an estimation on accuracy and variability. On the other hand, the 0.5° resolution (see comment later) may lead to the conclusion that the trajectories are not useful here at all.

lines 161 - 153 and Fig 2: I would not term the second particle class "OC-EC", because it contains a lot of nitrate and sulfate. Something like "OC-EC + inorganic" or "EC + secondary" would describe this type better.

lines 171 - 174: But you measure close to the Baltic. I would therefore expect to see also fresh sea salt particles at times when the air arrives directly from the north. Why are there no fresh sea salt particles?

Line 258: Why were only 12 out of 15 clusters used here?

Lines 274 - 279: The "transient group" is identified as ship plume particles. The question arises again: Would you get the same (or a similar) result without resonance-enhanced iron detection? What would happen if you use Vanadium as a marker ion?

Fig 4 b) What are weight matrices? To me the plots looks like "averaged mass spectra". Why do panels b) and c) have different sizes?

Lines 291 - 295 I don't agree that the absence of negative ions due to uptake of water is a sign of aging. It is more reflecting the actual humidity conditions and can therefore be a sampling issue. Drying the aerosol (intentionally by using a dryer or inadvertently by drawing cold air from the outside into a warm laboratory) will remove the water from the particles again. Stratospheric particles which have presumably spent a long time in the atmosphere show large  $\text{HSO}_4^-$  signals (e.g. Murphy et al., 2014).

Line 297: By "in the plume" here you mean gas plume inside the MS after laser ablation, not the ship plume, or am I wrong?

Line 313: "In contrast to negative ions and EC, OC as well as alkali cations". I suggest using consistently either "positive and negative ions" or "anions and cations".

Line 315-317: I think that is clear that Fe is enhanced. How large is the enhancement compared to non-resonant ionization? Is there an enhancement factor? In Passig et al. (2020) you report an increase by up to a factor of 20 for ambient data. But is that particle detection efficiency or signal height? This should be mentioned here.

Lines 331 - 334: Would it make more sense to take only the "transient" clusters from Fig 4 (110, 150, 151, 161,164, 183)? Or show these clusters in separate time series?

lines 354-369: I am certain that on such small local scales, wind direction is better than HYSPLIT with  $0.5^\circ$  resolution. The distance to the harbor was about 10 km, and  $0.5^\circ$  are in the Rostock region about 50 km in north-south direction.

line 357: Refer to Fig 5 here instead of Fig 4d. Add markers (numbers) to the plumes in Fig 5 and refer to those.

line 364-369: See comment to  $0.5^\circ$  degree resolution above. You can't expect to see such local features.

Lines 393 - 399: A more detailed plot of the time series for the individual plume would help a lot here.

Lines 434-435: I fully agree with this: local wind data and small scale models are absolutely needed.

lines 437-439: Multiple stations? I think that would be a huge effort for single-particle mass spectrometers. A measurement site closer to the shipping route but in the main wind direction (e.g. Diesch et al., 2013) would improve the approach already a lot.

### **Technical comments:**

Line 48: However, "of" scrubber ooperation? Remove "of"

Line 56 + 411: please correct citation "Van Roy, W and Scheldemann, K., 2016"

line 115 "if product" -> insert "the"

line 210: Insert full stop after S3

line 268 "his" -> "this"

### **References**

Ault, A. P., Gaston, C. J., Wang, Y., Dominguez, G., Thiemens, M. H., and Prather, K. A.: Characterization of the Single Particle Mixing State of Individual Ship Plume Events Measured at the Port of Los Angeles, *Environ. Sci. Technol.*, 44, 1954-1961, 10.1021/es902985h, 2010.

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