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Interactive comment on "Air quality monitoring in communities of the Canadian Arctic during the high shipping season with a focus on local and marine pollution" *by* A. A. Aliabadi et al.

Anonymous Referee #1

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Review of the paper "Air quality monitoring in communities of the Canadian Arctic during the high shipping season with a focus on local and marine pollution" by A. A. Aliabadi et al.

The paper describes the results of air quality measurements and their analysis at two locations in the Canadian Arctic during summer and autumn 2013. The measurements are of interest, because they describe the impact of shipping and other sources of air pollution in a, until now, mostly unpolluted region. However, the paper suffers from many doublings, imprecise evaluation methods and too speculative findings. It is far

too long in relation to its basis, which are two time series of observations with instruments which are only partly suited for environments with very low concentrations of air pollutants. The paper is not suitable for publication in ACP. It has to be rewritten in an extended way in order to make it readable and useful.

Major critics:

1. The given "estimated percent ship contribution to NO_x, O_3, SO_2 and $PM_{2.5}$" indicate that, on average, about 15-20 % of the measured concentrations of these pollutants stems from ships. However, if I understood the method presented in 3.4.5 correctly, this percentage is the enhancement of the concentrations in those cases when ships eventually contribute to elevated concentrations. There are three critical aspects in this method: 1) The identification of these cases is based on a trajectory analysis that uses coarse resolution (in time and space) meteorological data. It cannot be said which other sources of pollution may play a role and it cannot be said how likely it is that this individual ship, that is in the area of the trajectory, really contributes to the pollution levels in the sampled air mass. 2) A low number of events with high concentrations could give a high value in equation (7). In my understanding, some few events would not indicate a high contribution of shipping to air pollution, if the cases are rare. Therefore the measure presented here is not usable as an indicator for the contribution of shipping to air pollution. 3) Ozone formation largely depends on radiation and therefore exhibits a strong diurnal cycle. Following equation (6) the diurnal cycle would cause "pollution". A modified diurnal cycle for certain wind directions, which is caused by different meteorological conditions (which are likely connected to different wind directions) can cause changes in O_3 concentrations. These are then interpreted as changes in the pollution situation caused by certain emission sources although they are purely meteorological.

2. The measured concentrations, in particular for NO_x, SO_2 and $PM_{2.5}$ are on the

majority of the days just above the LDL, $PM_{2.5}$ is often even below it (in Fig. 9 and 10). The consequence is that $C_{10\%}$ in equation (5) is the LDL. The analysis is then based on a few pollution events, of which the majority is not from ships, according to Figs 9 and 10. To summarize this: I have severe doubts that the precision of the instruments is sufficient for this analysis. For the airpointer it is stated from the manufacturer that the precision is 1 ppb for NO_x and SO_2 and 1 $\mu g/m^3$ for $PM_{2.5}$. The majority of the NO_x and SO_2 values are around 1ppb or even below. The other concern that follows from this is that AQHI is almost entirely driven by ozone with concentrations between 20 and 50 ppb. Ozone concentrations are largely influenced by sources far from the measurement location and variations in its concentration may be caused by meteorological effects (as said above) or impacts from NO_x or VOC sources which are far away and cannot be identified.

3. The use of the AQHI as a measure for the deterioration of the air quality caused by shipping emissions is doubtful, because NO_x emissions from ships may lead to reduced ozone concentrations (depending on time of the day, availability of reaction partners and season) and therefore eventually lower AQHI values may result from air pollution by ships.

Other major comments:

page 7-9, section 2.2: The precision of the measurements is not given and it is not discussed whether it is sufficient for this analyses. You just give the variation of the zero air measurements in Table 1. My impression is that the precision is not sufficient. BC is in the text often given in $\mu g/m^3$ although it seems to be lower than 50 ng/m^3, according to Fig. 17.
page 10, section 2.4: All trajectories are based on 1 degree spatial resolution and 6 hr temporal resolution meteorological data. You interpolate them to 1 min resolution in order to make further use of them. I do not think this scientifically sound. Many regional and all local transport features that will influence the transport of pollutants in the Arctic

are not represented in this data, therefore the possibility to track ship plumes based on this data is very limited.
page 10: I do not see the use of the emission inventory for Canada North of 60 degrees. Many source regions, in particular for Cape Dorset are more south, as shown in Fig. 5. page 11, section 2.6: I commented earlier on this. The use of AQHI is not adequate to track small pollution events. page 12, section 3.1: It would be important to know how big these ships are and what their likely emission is compared to other sources.
page 13, lines 410-415: I do not have confidence in the usability of the trajectories for this analysis by looking at Fig. 6. The figure does not tell much. It seems that more than 50% of the cases have differences in wind direction which are above 30 degrees. I cannot judge whether this is sufficient. page 14, section 3.4: Why did you employ two clustering approaches for your analysis and not one more sophisticated one, if the algorithms exist? This makes the entire section 3.4 lengthy and difficult to understand. You end up with far too many plots and tables.
page 14, section 3.4.1: How can you be sure that you see ship events and not some other source of pollution which is in some distance from the measurement station. SO_2 should be a good tracer that indicates HFO burning, however, you do not see any elevated SO_2 concentrations (see lines 511-513 and Fig. 13). Why is this the case? As you state some pages before, there is no ECA in the Arctic, so ships presumably use sulfur rich HFO fuels.
page 16, lines 514-530: You describe the difficulties with AQHI in cases of ozone titration. For me, you need to conclude from this that it cannot be used to identify "polluted" situations.
page 16, section 3.4.3: I cannot follow the usefulness of this combined static and dynamic approach. Why don't you group the trajectories into those influenced by ships and those not influenced by ships. Of course you need to exclude all that are contaminated by local pollution beforehand.
page 17, line 567/568 and line 573/574: I do not think that the loss of particles due to

growth and deposition is a likely explanation. Secondary aerosols in the accumulation mode typically do not grow up to particle sizes that are subject to sedimentation.
page 18, section 3.4.4: Why are there less particles in the size range above 70 nm in the unpolluted cases in Fig. 19a? The explanations you give for the measured particle size distributions between lines 613 and 630 are very speculative and cannot be proven. In particular, if DMS is important, you should also see enhanced SO_2 levels.
page 19/20, section 3.4.5: The entire analysis presented in this section is obscure. See my comments above (Major critics, point 2).

Figures: There are too many figures. A number of figures obsolete, e.g. Fig. 3, Fig. 6, Fig. 11. Fig. 7 gives the same information as Fig. 8. Fig. 15 is hard to understand and of limited usefulness.
In Figure 14, 16, 17, 18: If the dot is the median what is then the inner line in the box and whisker plot? This should typically be the median.
Figures 16-18: I have severe doubts that you were able to detect ship plumes with your trajectory method. If I look at the numbers in Table 3, at Cape Dorset, you have 89582 trajectories with ship influence and 114491 without ship influence. This would mean that about 45 % of the arriving air masses are influenced by shipping emissions. Looking at Fig.9, one would expect a much lower fraction of air masses being influenced by ships.

Tables:
Table 1: This is the precision of the zero calibration. What about the precision of the measurements?
Table 2: I do not see the use of this information in this detail. It is never used.
Table 5: The units for EBC must be wrong.
Table 7: How many samples do you have for the unpolluted case?
Table 8 and 9: I have severe doubts that these numbers represent the fraction of pollution that is caused by shipping activities. See my comments above.

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