“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, R. M. Staebler, and S. Sharma

Science and Technology Branch, Environment Canada, 4905 Dufferin Street, Toronto, ON M3H 5T4, Canada

Response to Reviewers by A. A. Aliabadi on behalf of all co-authors

Dear Prof. Dr. Ralf Ebinghaus

We thank you and the reviewers for the constructive comments. We have addressed all issues in the responses below and will upload all necessary files to the portal, including the modified graphics, modified manuscript in LaTeX and PDF format [submission C], a highlighted version of the manuscript in PDF format, and two previous submissions [A and B] to communicate changes effectively and easily. Below you can find point-by-point responses to the reviewer comments. The comments are shown in *italics*, the responses are shown in plain text, and the modified text in the manuscript is shown in **bold face** with **“quotation”** marks. Here we refer to figures, tables, and line numbers associated with the original submission to the journal [submission A], the discussion phase paper [submission B: ACPD 14, 29547-29613], and the final revised paper [submission C].

It is our impression that reviewer 2 was generally satisfied with the methodology and conclusions but requested a revision by adding more references and explaining why AIS methodology was not used. We addressed all comments accordingly. Reviewer 1 had objections to the validity of the methodologies used and found the manuscript longer than it deserves. However, the nature of the detailed and useful comments indicates, implicitly, that a major revision is required. Unfortunately this reviewer used the very first manuscript submitted to the journal [submission A] and not the ACPD paper [submission B] for his/her comments. We had much improved the quality of the paper since submission A, as reflected in version B, which has probably not been seen by this reviewer. Nevertheless, we have provided evidence, to the best of our ability, that the methodology has merits in this application and that the results provide useful conclusions, although we acknowledge that there are limitations with our approach. Also, we have shortened the manuscript by removing 5 figures since submission A.

In conclusion we thank you for your time and trust your judgment in the final evaluation of this revised manuscript.

Best Regards,

Amir A. Aliabadi

Reviewer 1:

General Comments:

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.

Response: We thank the reviewer for providing detailed comments. We have shortened the paper by removing 5 graphs or tables. Below we provide specific responses in the criticisms about the methodology and speculations. We highlight the advantages and disadvantages of the methodology as well as our approach in interpretation of the analysis. We also clarify the basis for which the applicability of the methodology and interpretation of the results within the context may not have been fully appreciated by the readers.

Specific Comments:

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.*

   Response: The ship-influenced air masses in this calculation are not identified from "elevated concentrations", but they are identified using dynamic clustering: i.e. grouping air masses that cross ships, within a spatial tolerance, before arriving at the measurement location. The percent contribution is then calculated using equations 5 through 11 by the pollutant concentrations in these air masses subtracted from the 10^{th} or 50^{th} (for O3) percentile of the entire data set, taken as representative of unaffected conditions, This concentration difference is positive on average, resulting in a positive %

contribution. This is by merit of the calculation, and not selection of air masses with elevated concentrations. So the statement: "*this percentage is the enhancement of the concentrations in those cases when ships eventually contribute to elevated concentrations"* is not correct.

1) We understand that most trajectory models (HYSPLIT, FLEXPART, etc.) have coarse spatial and temporal resolutions. However, spatial scales associated with ship movement relative to that of air (i.e. "wind") are an order of magnitude smaller. In other words, average ship speeds are much slower than average wind speeds in the boundary layer. Also ship emission time scales (hours to days) are much longer compared to trajectory time resolutions (1-6hr). As a result, we think available trajectory models can resolve ship location within the trajectory time resolution (1hr~6hr) and spatial error (typically10- 20% of the trajectory length (Stohl et al. 1998 [cited in manuscript])). In other words, the success of trajectory models in identifying ship sources of pollution is similar to that of land based stationary sources of pollution. (*Note: this is contrary to spatial scales associated with aircraft movement. Aircraft speeds are an order of magnitude faster than the wind, so it is never possible to use the existing trajectory models in identifying aircraft pollution*). Other studies using the same trajectory model and resolution to identify pollution due to shipping traffic has been successfully published in ACP. Please see the paper "*Contribution of ship traffic to aerosol particle concentrations downwind of a major shipping lane" by Kivekäs et al., Atmos. Chem. Phys., 14, 8255-8267, 2014, doi:10.5194/acp-14-8255-2014*. In addition, figure 13 [in submission A or 12 in submission B] shows a very good correlation between ship-influenced air masses indicated by plume age using dynamic clustering, and the rise in pollutant concentrations (O_3 and $PM_{2.5}$).

Dynamic clustering of air masses is only sensitive to ship-influence. In other words, it is indifferent to other sources of pollution that could be in the vicinity of or concurrent with the ships. So this technique is a "sensitivity test" showing the effect of ship versus no-ship-influenced air masses "only". Our fig. 16-18 [in submission A or 14-16 in submission B] and table 5 successfully show a statistically significant (90% confidence) concentration difference (O_3, $PM_{2.5}$) between ship versus no-ship-influenced air masses. In fig. 16 [in submission A or 14 in submission B], the median, upper quartile, and the 90^{th} percentiles for O_3 and $PM_{2.5}$ are also above instrument lower detection limit. This is evidence that the dynamic clustering method is working and that ship pollution is detectable over any other possible background or concurrent pollution that may be in the vicinity.

2) We agree with this statement. High or low cumulative pollution due to ships, as calculated by the method of section 3.4.5, is not time result. So with this measure we do not know if the ship effect was persistent over time or if it was episodic, occurring at only a few instances. This measure is a cumulative indicator showing what fraction of the cumulative pollutant concentrations observed at the measurement location is likely to be due to ships. This is useful in trends analysis over, say, a 5-10 years period.

3) There is a weak diurnal cycle for solar radiation in the summer-time Arctic because the days are 24hr long. We have performed a Fast Fourier Transform spectral analysis and found that the diurnal variation in O_3 concentration is negligible compared to effects such as long range transport, titration, and photochemical enhancement at larger or smaller frequencies than 1-day. The only other significant natural process governing O_3 concentrations in the Arctic is halogen chemistry, which mainly occurs in the polar

sunrise (spring), so it does not affect O_3 variations in our analysis period. So we were not concerned with diurnal ozone variations artificially being interpreted as pollution.

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 C10% 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 µg/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.*

   Response: Figs. 9 and 10 [in submission A or 8 and 9 in submission B] only demonstrate pollution time series in the 25-75 percentile values of the daily measurements. Since the Arctic is a clean and pristine environment, high pollution peaks are short-lived and end up in the 75-100 percentiles. So they do not appear in these figures. To shorten the paper length and remove this ambiguity, we have decided to remove these figures. On the other hand, the time series in fig. 13 [in submission A or 12 in submission B] show peaks as high as 6-10 µg/m^3 for $PM_{2.5}$ and 1.8 ppb for NO_x due to ship pollution and much higher values for NO_x, SO_2 and $PM_{2.5}$ due to local pollution. Also the box plots in figure 16 [in submission A or 14 in submission B] clearly show the median, 75th, and 90th percentiles well above LDL for most species since these air masses are grouped for specific sources of pollution. Having said this, we already acknowledge in the paper that higher NO_x, and SO_2 concentrations result from local emissions and not from ships due to LDL not being low enough. The figs. 9 and 10 [in submission A or 8 and 9 in submission B] were not supposed to demonstrate pollution due to shipping. We used dynamic clustering of air masses for this purpose.

   It is true that AQHI time series is dominated by long-range O_3 concentration. However, shipping influence on AQHI is successfully demonstrated in a statistically significant way using the dynamic clustering approach in the fig. 18 [in submission A or 16 in submission B] and table 6. It is already acknowledged in the paper that the shipping influence on AQHI is less than the influence from long-range O_3, but it can be quantified. Specifically in section 4 (Conclusions and future work) on page 31 [submission C] we state:

   "The high resolution Air Quality Health Index (AQHI) primarily followed seasonal O_3 levels and was higher for Cape Dorset than Resolute. Ship-influenced air masses consistently exhibited an increase of 0.1-0.3 in AQHI compared to no ship-influenced air masses. This difference is small with existing low levels of shipping traffic in the Arctic, but it can be expected to intensify with increasing traffic."

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.*

Response: As suggested in the previous comment, we were able to show increased AQHI due to shipping using the dynamic clustering approach. However, we fully agree with not using AQHI in performing detailed health risk assessments for other reasons: AQHI in this study is used as a simple instrument to put the results in perspective. Our personal communications with Health Canada have resulted in deciding to provide other clarifying statements regarding the limitations of AQHI usage and introducing more advanced air quality health risk models such as AQBAT. Detailed assessments of health risk in this study, however, was beyond the scope of our analysis. Please see full response to comment 4 by reviewer 2.

4. *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 analysis. 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 μg/m^3 although it seems to be lower than 50 ng/m^3, according to Fig. 17.*

   Response: In the authors' opinion, the prescribed method for measuring precision is appropriate. During the internal zero air calibration, zero concentration of the pollutants are supplied to the analyzers. Any remaining variation in the measurement is a realistic estimate of instrument precision under field conditions. This "real-time" measurement in the field is preferred over reliance on manufacturer specifications only. The subsequent results in tables 4 and 5 are made in bold face if they are above the instrument precision. To improve clarity, the following statement has been added to the manuscript in section 2.2.1 (Gaseous pollution and $PM_{2.5}$ measurements) on page 12 [submission C]:

   "Subsequent tabulated measurements of concentration differences are indicated with bold face if they are above the instrument precision."

   The units for BC has been corrected already in submission B.

5. *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.*

   Response: we agree with this statement. We understand that trajectories have low spatial and temporal resolution and their use may be limited. However, since our goal was to assess the overall impact of shipping on air quality over a period of 5 months, we could not afford, computationally, any other high resolution model. For example, plume dispersion modeling would require at least hourly runs from all ships for the entire shipping season. This demands immense computational budget and produces very large datasets, the results of which are not guaranteed to be significantly different from ours. Such approach would be more suited for a single ship plume measurement. The authors are in fact involved in another study to measure and quantify a single ship plume in an aircraft study in the Arctic during the summer of 2014. This limitation has been acknowledged in section 4 (Conclusions and future work) on pages 31-32 [submission

C].

6. *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.*

   Response: The trajectory frequency plots shown in Fig. 5 are based on 120-hr (5-day) backward time. However, the analysis is performed for 16-hr, 24-hr, and 72-hr backward time. So it is true that for many cases the industrial pollution sources from the south, as indicated in this fig., are located outside of the trajectory coverage. The point of the emission inventory data was to put our results in context, so we would prefer to keep the associated figure and table.

7. *Page 11, section 2.6: I commented earlier on this. The use of AQHI is not adequate to track small pollution events.*

   Response: Please see response to comment 3 above, and full response to comment 4 by reviewer 2.

8. *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.*

   Response: In the manuscript, we have added a reference to the ship types and maximum engine power that travelled through the Arctic region and also the vicinity of each site. Both Heavy Fuel Oil (HFO) and distillate fuels are burnt by these vessels. The revised manuscript in section 2.1 (Vessel traffic dataset) on page 11 [submission C] now reads:

   "In total, 109 ships (with Lloyd registration numbers) were active in the Canadian Arctic during the 2013 shipping season. These ships were merchant, passenger, cargo, fishing, tug, cruise, coast guard icebreaker, and other types ranging from ~2000 HP tugs to ~30,000 HP coast guard icebreakers."

   Elsewhere in section 3.1 (Vessel traffic patterns) on page 19 [submission C] we have revised the manuscript:

   "Traffic near Resolute, however, appears denser compared to Cape Dorset. We have observed a maximum of 5 ships at a time (total of 22) within the vicinity of Resolute, compared to only 3 ships at a time (total of 39) for Cape Dorset (not shown)."

   The following statement is revised in section 3.4.5 (Contribution of shipping and other sources to cumulative pollution) on page 30 [submission C]:

   "Some frequent vessels in the Canadian Arctic, such as the Amundsen coast guard icebreaker, burn distillate fuels as opposed to heavy fuel oil."

   Despite these clarifications, the ship pollution characteristics are not resolved as a function of vessel type in this study due to limited scope and other reasons (the full justification for this decision is given in the response to comment 1 from reviewer 2). As a result, providing the detailed list of specifications for all 109 ships was beyond the

scope of this manuscript.

9. *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.*

   Response: The authors agree that fig. 6 [submission A] does not tell much in the sense that it does not directly prove or disprove suitability of the trajectory clustering method in our application. We believe that the results of figs. 13, 16-18 [in submission A or 12, 14-16 in submission B], and table 5 are better indicators that the method, although limited, has worked in identifying ship pollution. As a result we have eliminated this figure and the associated discussion.

10. *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.*

    Response: The authors are not aware of commonly used algorithms for trajectory clustering associated with moving sources of pollution. All the available algorithms that we have encountered (e.g. TSV, k means, BIRCH, DBSCAN, etc…) group air masses based on fixed geographical regions. To track ship influenced air masses, we had to develop the dynamic clustering approach allowing for moving sources (ships), while the static clustering approach is similar to the other commonly used algorithms. We believe that the two clustering approaches are complementary to each other. The static clustering approach makes possible distinguishing pollution that is coming from fixed geographical origins (e.g. lower versus higher latitudes, or ocean versus nearby town). On the other hand, the dynamic cluster distinguishes pollution in ship-influenced air masses.

11. *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.*

    Response: As stated in comment 1.1, dynamic clustering is insensitive to other sources of pollution since these will contribute, in a random fashion, equally to non-ship and ship trajectories. Therefore, on average, the difference demonstrates the sensitivity of concentrations as a function of ship influence. The fact that figure 16-18 [in submission A and 14-16 in submission B] and table 5 consistently show higher O_3 and $PM_{2.5}$ concentrations for ship influenced air masses is evidence that the influence is from ships. Given the sparsity of other pollution sources within a 3-day trajectory radius (Fig.5), it is physically unlikely that other sources of pollution are exactly aligned with individual ship trajectories.

    Even though the Arctic is not under an ECA, it is still regulated by MARPOL; ships are required to burn fuel with less than 3.5% sulphur content. In addition, frequent coast guard icebreakers (such as the Amundsen) burn distillate fuels with lower sulphur

contents. Nevertheless, under section 4 (conclusions) we have stated that SO_2 pollution events DO occur due to ships, but rarely and episodically, so that no statistically significant statement can be made regarding the effect of shipping on this pollutant. We have also acknowledged that the lower detection limit of the instrument is not low enough to measure weak SO_2 signals from ships.

12. *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.*

    Response: We had already removed this discussion and figure 15 [submission A] from the manuscript in submission B. We have avoided using AQHI statistics, due to its limitations, in the static clustering analysis. For dynamic clustering analysis, as suggested earlier, pollution sources that titrate ozone will contribute, in a random fashion, equally to ship and non-ship trajectories on average. Our approach is a "sensitivity test", only showing the separated effect of shipping on AQHI.

13. *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.*

    Response: The dynamic clustering technique performs exactly the type of grouping suggested. The authors are aware of common techniques, such as FFT frequency filters or other spectral methods, in removing local pollution from long-range pollution. However, shipping exhibits both features. Nearby ships "appear" as local pollution with sharp and narrow peaks, while distant ships "appear" with low and wide peaks. Neither did we have the capacity to measure chemical components of pollution (mass spectrometry) to clearly identify ship pollution by tracers. So in this application, we could not eliminate local pollution easily, and had to resort to sector analysis (A, B, C, and D) to provide some useful statistics. Readers may acknowledge the difficulty of such remote measurements in the harsh environment of the high Arctic. While we believe that the dynamic clustering is the best approach, within the constraints of this project, for separating out the contribution of ships to local pollution, the static clustering approach provides the necessary context for the reader to assess the totality of the observations at Cape Dorset and Resolute as a function of trajectory sector. Building on this context, the dynamic clustering approach then separates out cases with and without ships upwind.

14. *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.*

    Response: Thank you, we have removed this statement.

15. *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.*

    Response: Fig. 19 [in submission A or 17 in submission B] showed a number "fraction" distribution of particle size and not the "absolute" number concentration. i.e. the area under the curve for each distribution is the unity. So the absolute magnitude of the mode

at one particle size cannot be directly compared among different curves (distributions) associated with different plume ages or background conditions. Only the relative magnitude of the multiple modes for a “single” curve (distribution) in these plots has physical meaning. Of course, absolute number concentrations for fresh plumes are much higher than the background in most particle sizes, but this does not appear in the number “fraction” plots.

We have acknowledged in the first sentence of this section that our conclusions in this section is speculative and limited, but they still reveal important differences in particle size distribution between ship polluted air masses and non-ship polluted air masses.

16. *Page 19/20, section 3.4.5: The entire analysis presented in this section is obscure. See my comments above (Major critics, point 2).*

    Response: Please see response to comment 1.2, the indicators calculated in this section are cumulative. These indicators simply combine the air mass movements and the annual ship traffic patterns to give an estimate of the contribution of ships to locally observed pollution levels. This indicator is useful in long-term trend analysis in a, say, 5-10 year time period.

17. *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.*

    Response: The reviewer suggestion was taken into consideration by removing 5 less important figures from the paper since submission A. These are figs. 6, 7, 9, 10, and 15 [submission A]. The dot is the median for the “entire” dataset, while the line is the median for the specific grouped sample. Thus, the 4 median dots for O_3 for each location are the same. Some grouped samples exhibit a higher or lower median than the median for the entire dataset.

    Again, in submission B, the episodic time series analysis (fig. 12), dynamic clustering technique (figs. 14-16), and the particle size distributions (fig. 17), all provide evidence that the ship plume could be detected using the trajectory method.

    Please note that the analysis period is limited to the shipping season and that Cape Dorset is located south of Baffin Island close to the Arctic Bridge shipping corridor. In the shipping season many Arctic communities receive their annual resupply via the Arctic Bridge. Also there is active traffic associated with mine development and logistics in this corridor. Given these, the estimate of 45% influenced air masses is not surprising. The estimation of this statistic for the entire year will be much lower.

    As suggested in response to comment 2, figs. 9 and 10 [in submission A or 8 and 9 in submission B] did not show pollution in the daily 75-100 percentiles, where shipping

pollution is expected, so they have been removed from the manuscript.

18. *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.*

    Response: The only precisions that could be calculated in the instrument were for the zero and span. Most ship pollution concentrations are closer to the instrument internal zero than they are to the span.

    Table 2 puts our shipping pollution results in context of local pollution estimates. The units for EBC in table 5 was already corrected in submission B. In table 7, the SMPS output was every 9 minutes. So given a period of 5 months the total number of samples was ~24,000. The number of polluted samples was 904 or 3.8% of the entire sample size.

    Tables 8 and 9 are only associated with the shipping season. If these statistics are calculated for the entire year, the % contributions will be much less, especially for Cape Dorset.

    These estimates and those shown in tables 5 are in alignment with those reported in the literature. For example, Granier et al. (2006) and references within [cited in manuscript] suggest a current level ~4ppb increase in surface level ozone due to Arctic shipping. Our table 5 reports 2.7-2.9, 4.3-4.5, 4.6-4.7, and 2.5-2.7ppb increase in ozone due to shipping depending on site and wind sector. Browse et al. (2013) [cited in manuscript] suggest a 1-15% (their figure 2) BC deposition due to shipping in the Arctic, depending on location. Our estimate in table 8 for surface level BC due to shipping is 4.3-9.8%. We find these agreements very encouraging.

Reviewer 2:

General Comments:

The manuscript of Aliabadi, Staebler and Sharma "Air quality monitoring in communities of the Canadian Arctic during the high shipping season with a focus on local and marine pollution" touches the timely discussion of ship emissions in the northern latitudes. The paper describes the measurement methodology and source apportionment of air quality observations with the help of modeling tools. I find the topic very interesting and the manuscript well structured. However, the authors have chosen to neglect some relevant data sources, like AIS data, which seems strange, especially since the authors cooperate with maritime authorities who can provide this data.

Response: We thank the reviewer for providing detailed comments. Reference to AIS data, its advantages and disadvantages, and justification for its exclusion from the analysis is provided in detailed comment #2 below.

Specific Comments:

1. *Chapter 2.1. Here it is stated that the authors are using data from Canadian Coast Guard, which includes Lloyds registration numbers. However, in the Conclusions and future work section (page 29578, lines 25-27) it is stated that no data for ship engines were available. It is not stated whether data for individual vessels for example from IHS Fairplay were used or not. It should be noted that the classification societies' websites (DNV GL Exchange, Americal Bureau of Shipping, Korean Register, Russian ship register, Class Nippon Kaiji Kyokai, Bureau Veritas) offer this data for free for individual vessels (searchable by IMO number). This makes the statement "engine data was not available" a bit flimsy and the authors should explain why they have not used this approach.*

   Response: Thank you for introducing the new sources for obtaining ship engine information given ship IMO number. We have made a reference to this in the manuscript. However, the detailed analysis of emissions characteristics as a function of ship type, engine size, fuel, and engine load was not done for the following reasons:

   First, the scope of this paper was to quantify the relative impact of shipping emissions, all engine types, sizes, and fuels combined, versus that of local pollution and long-range continental transfer during the entire shipping season. Emissions characteristics as a function of engine type, size, and load is dealt with in detail in the literature using a complete suite of instruments in plume intercept, stack measurements, or test-rig studies. Please see papers by Jalkanen et al. (2009), Petzold et al. (2010), Petzold et al. (2011), Lack et al. (2011), and Cappa et al. (2014) as referenced in the manuscript. Second, even if we did attempt to resolve emissions characteristics as a function of engine, size, type, and fuel, we would not be able to perform a complete analysis. We would still lack information on engine load as it is not logged in data sources suggested. Also we would suffer from poor statistics and small sample sizes for each ship plume encountered, as inferred from table 3 [submission C]. So per request we have revised the manuscript as the following: Section 3.4.4 (Particle size distributions) on page 26 and Section 4 (Conclusions and future work) on page 31-32 [submission C] now read:

   "These results do not resolve variability in engine type, engine load, and fuel type due to the limited scope of the analysis and the limited number of samples available for each ship plume encountered. However, information on individual engine types, sizes, and fuels are available from classification societies such as IHS Fairplay, DNV GL Exchange, American Bureau of Shipping, Korean Register, Russian ship register, ClassNK, Bureau Veritas."

   "Plume dispersion modeling for our purposes was virtually impractical since it would have required plume dispersion simulations for each ship at short time intervals for the entire shipping season, which was an immense computational calculation and beyond the scope of our analysis."

2. *Section 2.1: It seems that Automatic Identification System data was not used for this work. Why not? At least it should be mentioned that AIS data could be used for this purpose. It seems that not all ship activity data sources were included and the authors should explain why they have not done so.*

Response: The authors are aware of AIS data, its advantages and disadvantages, and its previous use in the Arctic region. AIS data is obtained by ships equipped with transponders that communicate ship location to either land (terrestrial) or satellite receivers. AIS ship communication in the European Arctic has been reported at a frequency of every 2 hours or better by Winther et al. (2014) "Emission inventories for ships in the Arctic based on satellite sampled AIS data" (Atmospheric Environment).

We did not use AIS for the following reasons: The Canadian Coast Guard (CCG) authority does not provide AIS data to government agencies in Canada free of charge. The Canadian Arctic is not equipped with any land receivers and any AIS ship traffic in the region is solely based on satellite observations. Given that since not all ships are equipped with transponders, the AIS data will not contain all ships but provides very high resolution for ship locations identified in its database. On the other hand, the CCG database, which is based on activity reporting, has ships with more than 98% compliance in activity reporting, but the temporal and spatial resolutions are not as high as any AIS data. Additionally, given the low level of resolution and accuracy associated with air mass trajectory models, we believe that the CCG database is sufficient for our purpose of estimating relative ship impact on air quality versus other sources. Nevertheless, we have added the following statement in the manuscript to introduce AIS as an alternative for ship traffic tracking in Section 2.1 (Vessel traffic dataset) on page 10 [submission C]:

"Alternative to the Canadian Coast Guard dataset, the Automatic Identification System (AIS) could have been used in ship traffic tracking with greater temporal and spatial resolution but possibly fewer ships identified. This technology has been successfully used over the European Arctic in tracking ships (Winther et al., 2014). For the purpose of our analysis, however, we believe that the Canadian Coast Guard dataset is sufficient since, given the low resolution and accuracy associated with air mass trajectory models, these datasets are not expected to give different results."

3. *Section 2.5: To my knowledge, tire and brake wear do not contribute to SOx pollution. Usual markers for tire and brake wear are Fe, Cu, Sb and Ba whereas resuspended mineral dust (sand) consists of Ca, Si, Fe and Al components. Table 2 seems to list tire & brake wear as sources for SOx, which is odd.*

   Response: The main source for this table is the National Pollutant Release Inventory (NPRI) available at http://www.ec.gc.ca/inrp-npri/. The authors did not contribute in the NPRI dataset. Specific questions can be directed to the site under "Contact" menu. We did not have the capacity to measure any of the trace elements mentioned at these sites (i.e. Fe, Cu, Sb, Ba, Ca, Si, and Al).

4. *Section 2.6, AQHI use: Equation (1) does not seem to describe polyaromatic hydrocarbons. Are these included in health effect evaluation? Also, in Stieb et al (2008), AQHI goes to zero whereas here the authors say that the minimum is one. Why?*

   Response: AQHI can physically and mathematically go to zero. However, operationally it is rounded to an integer with a conventional arbitrary minimum value of 1. The following sentence has been revised in the manuscript in Section 2.6 (Air Quality Health Index

(AQHI)) on page 17 [submission C]:

“Operationally, AQHI is rounded to the closest integer with an arbitrary minimum value of 1.”

AQHI in this study is used as a simple instrument to put the results in perspective. This tool does not account for health impacts of polyaromatic and hydrocarbons. Our personal communications with Health Canada has resulted in deciding to provide other clarifying statements regarding the limitations of AQHI usage. Our colleagues in Health Canada have suggested that if a more detailed and accurate health impact assessment of shipping pollution is desired, a more advanced and statistical health model, such as the Air Quality Benefits Assessment Tool (AQBAT), is necessary. This is, however, beyond the scope of our analysis, which is primarily focused on air quality monitoring research. The following statements are revised in the manuscript in Section 2.6 (Air Quality Health Index (AQHI)) on page 17-18 [submission C]:

“AQHI is used as a simple tool to put our results in context. Assumptions pertaining to threshold or no-threshold formulations, effect of long term background pollutants, smoking habits, and other various factors have long been debated in the formulation and output of air quality health models. If a more detailed and accurate health impact assessment of shipping pollution in Arctic communities is desired, a more advanced and statistical health model, such as the Air Quality Benefits Assessment Tool (AQBAT), should be used. Such a model enables the definition of various specific scenario models from the flexibility of combining and linking of pollutants, health endpoints, geographic areas, and scenario years (Judek et al. 2012).”

5. *Section 3.1: Here references to Figs 8 and 9 are made, but Figs 8-9 are introduced later, in Section 3.2.2. Could the authors check the consecutive numbering of figures?*

   Response: Thank you. We have decided to remove figs. 8 and 9 [submission B] since there is a level of redundancy in other figures. This helps shortening the paper.

6. *Section 3.4.1: Authors state that sometimes the local pollution events are superimposed with the ship plumes. Looking at Figs 7 and 12a and 12c, this seems to occur 2/3 times in Cape Dorset. In Fig 12a, the wind blows from west, which is marked as "Town" and "Waste burn" sectors in Fig 7. How can the ship plume be identified in these cases, if the wind blows away from the measurement site? Fig 12b seems to be consistent with the wind direction, though.*

   Response: The readers of the paper must be careful to fully understand grouping of air mass samples based on static and dynamic trajectory and local wind directions. The town pollution arrives at monitoring sites from ‘*stationary*’ sources, while the ship pollution arrives from ‘*moving*’ sources. In addition, the town and ship pollution signals are not completely ‘*mutually exclusive or inclusive*’ since both sources can be intermittent, meaning that the signals may sometimes be superimposed while at other times they may be separate. The signals cannot be fully separated using our methodology or frequency decomposition due the nature of the ship signal since some ship signals from the vicinity of the town may appear as town pollution while at further

distance the signals exhibit a different time signature. However, given the logistics of the town emission sources and site wind direction, it is possible to infer which wind directions bring *'almost pure*' ship pollution and which wind directions bring ship signals '*possibly contaminated*' by town pollution. Dynamic clustering of trajectories (based on ship and no-ship influence) and local wind directions (A+C, and A+B+C+D) enable calculating pollution concentrations in 4 sub-groups: with and without ship influence as well as with and without town influence. These sub-groups further allow estimation of an upper and lower range for the cumulative ship impact versus other sources.

Fig. 12a [submission B] shows a ship plume from the west of the Cape Dorset peninsula. The wind from the 270 degrees direction brings the ship plume to the monitoring site, but the air mass picks up town emissions on its way, as indicated by sharp peaks and supported by Fig. 7. In Fig. 12b wind is blowing from the east of the Cape Dorset peninsula, with less influence of the town emissions. In Fig. 12c the situation is similar to that of Fig. 12a. This demonstrates that the ship and town signals are not completely '*mutually exclusive or inclusive*'. In these instances, roughly ½, 1/5, and 2/3 of the ship signal was contaminated by town emissions, associated with Fig. 12a, 12b, and 12c.

7. *Section 3.4.3, lines 575-578. Authors state that "shipping degrades the air quality, in a statistically significant way, with current traffic levels". However, it should also be mentioned how the findings relate to WHO limits for PM, NOx and SOx.*

Response: Thank you. After the discussion in comment 4, we decided that it is wiser to leave out the statement regarding '*degradation of air quality'*. We can only report a statistically significant change in AQHI, but should leave any accurate or detailed statements about health effects of ship pollution to more advanced health models and the health authorities. Section 3.4.3 (Dynamic clustering of air mass trajectories based on ship presence upstream) on page 25 [submission C] now reads:

"The mean difference in AQHI between ship and no ship-influenced air masses varies from 0.1 to 0.3, in a statistically significant way, with current traffic levels."

8. *Section 3.4.4: Authors state that DMS contributes to sulphate/PM measurements, but do not mention sea salt in this regard. According to O'Dowd et al (Atm. Env., 31, 1997, 73-80) this would seem relevant in the size range reported by the authors. Was sea salt contribution considered in the analysis?*

Response: Figure 17 [submission B] shows that a large accumulation mode persists in the aged air mass, likely with some influence from primary sea salt aerosols. We do not have any chemical measurements to support this, but the influence is implicitly present in the particle size distribution. We have added the following statement to section 3.4.4 (Particle size distribution) on page 28 [submission C]:

"The accumulation mode may also be influenced by ocean-originated sea salt, as suggested by O'Dowd et al. (1997)."

9. *Section 3.4.5, lines 665-671. In Resolute, BC measurements were made. Is it possible that the vessel plumes are produced from burning distillate fuels instead of HFO? This could explain why BC emissions are lower than found in the existing literature. Did the*

Resolute measurement setup include measurements for Ni or V, which are usually considered as markers of HFO burning?

Response: Thank you. In fact we know that some frequent vessels in the Canadian Arctic do burn distillate fuels, such as the Canadian Coast Guard icebreakers (e.g. Amundsen). So it is likely that this may be responsible for lower BC fractions in PM2.5. We did not measure Ni or V, which are markers of HFO burning. The following statement is revised in section 3.4.5 (Contribution of shipping and other sources to cumulative pollution) on page 30 [submission C]:

"Some frequent vessels in the Canadian Arctic, such as the Amundsen Coast Guard icebreaker, burn distillate fuels as opposed to heavy fuel oil. In addition, we suggest that after aerosol processing over many hours (our case), hydrophilic components of total particulate emissions grow, acquiring more mass, but the EBC components (mainly hydrophobic) do not. These facts justify our slightly lower mass fractions of EBC at the site."

10. *Figure 7. I would recommend arranging the labels in such a way that they do not overlap with each other.*

    Response: Thank you. This has been implemented.

11. *Figure 10. Is there a contribution from residential heating (small scale wood combustion) in PM results? There seems to be a slight upward trend in PM concentrations towards to the end of the study period. Levoglucosan can be used to trace this contribution. Would it be possible to identify the relevance of small scale wood combustion from the PM results?*

    Response: We know for a fact that wood combustion is a possibility, especially as we have observed it in waste burning of old furniture in Resolute. However, to our knowledge Diesel fuel is the primary source of heating energy for residences. We did not have the capability of measuring Levoglucosan. The upward trend in $PM_{2.5}$ measurement in Resolute towards the end of the study period may be caused by long-range transport as well as local combustion.