Response to Reviewer #1 comments

For clarity, our response to the reviewer's comment is in ***purple*** *font.*

This paper reports on the long term (2003-2020) measurements of atmospheric trace gases and aerosols from an island in Finland, with a particular focus on the impact of shipping regulation on SO2. The authors show that the SO2 concentrations have decreased significantly, especially since 2015, while NOx, PM2.5 haven't really changed. This is clearly a very valuable timeseries dataset, thanks to its length. However at present I don't feel like I've learned much new from reading this paper. There have been earlier studies that report on the reductions in atmospheric SO2 in coastal areas following the IMO regulations, which this paper fails to acknowledge. Other questions that could be (but not currently) addressed this paper include:

- Fuel sulfur content and the rate of compliance by ships
- Does the observed trend in SO2 reflect what one expects (e.g. based on atmospheric transport modelling with STEAM emission and terrestrial S sources)?
- Atmospheric processing of trace gases

Reply: 1) We would like to thank the reviewer for his comment. However, as we do not have simultaneous CO_2 measurements next to SO_2 measurements, we were unfortunately not able to analyze fuel sulfur content and the rate of compliance with legislation implemented.
2) Figure 6 presents the observed SO_2 to NO concentration ratios, demonstrating a significant reduction in SO_2 levels following the implementation of SECA regulations in 2015. This decreasing trend aligns with Figure 3, which shows a clear downward trajectory in SO_x emissions not only from the shipping sector but also across various other sectors (e.g., public power, industry) in countries bordering the Baltic Sea, including Finland and, consequently, the Utö site. Additionally, this trend is consistent with the predicted SO_x emissions from maritime traffic, as shown by the STEAM model in Figure 2.
Moreover, while employing a chemical transport modelling would be useful, it's important to note that this was not within the scope of this study. However, we understand the importance of such modeling in interpreting our findings, thus our data is available for those who wish to conduct this kind of analysis.
3) Regarding point 3, we would like to point out that the shipping lane is very close to the measurement location and the emissions reach the measurement location in under 10 minutes (according to now former Table 3). Additionally, considering that atmospheric conditions such as O_3 levels and solar radiation have remained relatively stable over the past two decades, we believe that atmospheric processing doesn't have a major impact on SO_2 concentrations, and observed changes in close-by plumes we measured.

Specific comments

Line 66. This paragraph is ok in isolation. But it's not something this work will address. It's probably better to remove or significantly shorten it.
Instead, before line 79 it would be useful to review previous work on ship SO2 emissions (including time series measurements similar to this study, e.g. doi:10.5194/acp-15-5229-2015 and http://www.atmos-chem-phys.net/16/4771/2016/). What are the knowledge gaps that this paper can fill? Is it e.g.
- impact of regulation on atmospheric pollutant level?
- scrubber vs. low sulfur fuel?
- rate of compliance from ships?
- attribution of emission to different ship types?
- atmospheric processing and transformation of trace gases

Reply: Thank you for the helpful feedback. Regarding the paragraph at line 66, our goal was to underscore that while the SECA regulations were primarily introduced to protect human health by reducing the sulfur content in fuel, they've also had some unintended consequences on climate and marine ecosystems. However, we have significantly shortened the paragraph and moved it to a more appropriate place within the introduction section to improve the flow and the transition from one paragraph to the next. We also added the papers the reviewer suggested (Yang et al., 2016; Beecken et al., 2015), including three more references. The introduction and the references sections were, therefore, revised.
We also feel that our study fills an important gap by providing a detailed, long-term analysis of air quality trends in the Baltic Sea, since to our knowledge, there are no other published long-term SO_2 datasets that have been collected in the middle of the sea for the Baltic Sea. We hope this work, together with the dataset described in this paper, and made available will contribute valuable insights into the effectiveness of SECA regulations and serve as a benchmark for local and regional scale dispersion modeling.
We have revised accordingly the Conclusions section to address the knowledge gaps that this work fills in and explain its novelty in comparison to similar studies.

4.1 there is nothing wrong with this section on its own. However I don't feel like it contributes very much to the paper at the moment. I guess the key message is that S emission from non-shipping sectors has been declining gradually, while S emission from shipping has decreased in step wise fashion following the IMO regulations (which we would've expected even without STEAM model)?
I think this section can be made more powerful if the authors implement these emissions in an atmospheric transport model, see what the predicted change in SO2 is, and then compare the model with the observations.
Reply: We thank the reviewer for his suggestion. However, the focus of this paper is on observed time series with some supporting data. Hence, employing an atmospheric transport model is outside of this study's scope. As the data will be available for modelers, we are really interested in seeing in the future outcomes of the modeling studies utilizing our observations

Line 158. SO2 lifetime was estimated to be only 0.5 day to the west of the UK. http://www.atmos-chem-phys.net/16/4771/2016/ in cloud oxidation is probably the largest sink.
Reply: Lee et al. (2011) and Beirle et al. (2014) evidenced that the average lifetime of SO2 in the lower atmosphere is ~1–3 days. However, this can vary according to atmospheric conditions, temperature and humidity. The duration of SO_2 lifetime doesn't have an impact on the SECA regulations. The above-mentioned references (i.e. Lee et al. (2011) and Beirle et al. (2014)) were added to this line to support the statement as well as to the references section.

Fig.2 how come the SOx emission from shipping sector hasn't decreased in step-wise fashion, corresponding to the regulations? Is it because the emission also includes outside of SECA?
Reply: We would like to point out to the reviewer that the y-axis in Figure 2 is on a logarithmic scale, thus making the gradual reductions in SO_x emissions probably less visually apparent. Moreover, as it is stated in the figure caption, Figure 2 demonstrates the combined annual SO_x emissions from Estonia, Finland, Lithuania, Latvia, Sweden, Russia, and Poland, all of which are within the SECA. Therefore, it does not include emissions from outside the SECA, ensuring that the data reflects the impact of SECA regulations alone. However, Figure 2 was revised to enhance its resolution and the following statement "*Note that the figure is presented on a logarithmic scale (y-axis). The source of SOx emissions data for the different sectors is EMEP (European Monitoring and Evaluation Programme)*." was added to the figure caption.
In addition, Figure 3 shows clearly the step-wise reductions of SO_2 following the introduction of each SECA regulation.

Table 1. a bit more detail on the SO2 measurements would be useful (even if this info had been reported previously elsewhere). E.g. how was it blanked and calibrated?

Reply: Per reviewer's request, we added information on how instruments were blanked and calibrated to subsection 3.1, below Table 1.

Line 169-170 this is a repeat

Reply: We would like to thank the reviewer for his observation. We have deleted this sentence as it was repeated earlier.

Line 176. 'until'

Reply: We replaced "till" with "until" in the manuscript.

Table 2. what's N(%)?

Reply: N (%) represents the fraction of the year for which high-quality data is available, expressed as a percentage. High-quality data, used in our analysis, are defined as valid data recorded, excluding those compromised by factors such as instrument malfunctions, environmental interference, or calibration issues. We have added the definition of N (%) to the caption of Table 2 and Tables A1-A4.

Figure 5. how come percentiles and median are not shown for data over the first few years? Is it because the data were hourly, not minutely? In general, I don't really see the value of presenting/analyzing minutely data for this section. Hourly data would be perfectly fine for looking at long term trends. Minutely data contain much more measurement noise, especially for SO2.

Reply: We would like to clarify that for the first period the data is only hourly and, thus, 1st and 99th percentiles for this period are not comparable with the second period with 1-minute resolution data. Analysis of nearby plumes requires higher time resolution data since typically the plumes from the ships are few minutes in duration; therefore, we have employed 1 minute data. In addition, 1-minute data is also better suited for possible later local and regional transport modelling studies.

Line 238. How were 'peaks' identified/defined? What's the minimum NO concentration in this calculation? Have you accounted for any possible lag between the SO2 and NO data due to imperfect time synchronization or different instrument response times?

Reply: Peaks were identified as a sudden and simultaneous increase of NO and SO_2 concentrations. The minimum peak prominence was set to be 2 μg m^{-3}. The different instrument response times have not been taken into account since the difference is at maximum few seconds and the data is presented in time resolution of 1 minute. The peaks in the data caused by a ship passing by Utö during wind blowing from the shipping lane towards the measurement station are typically very clear and well synchronized (see the figure below presenting a plume by a ship, not included in the manuscript). The analysis shows qualitatively that the SO_2 peaks caused by near-by-sources (ships) are significantly lower after SECA2015 compared to earlier years.

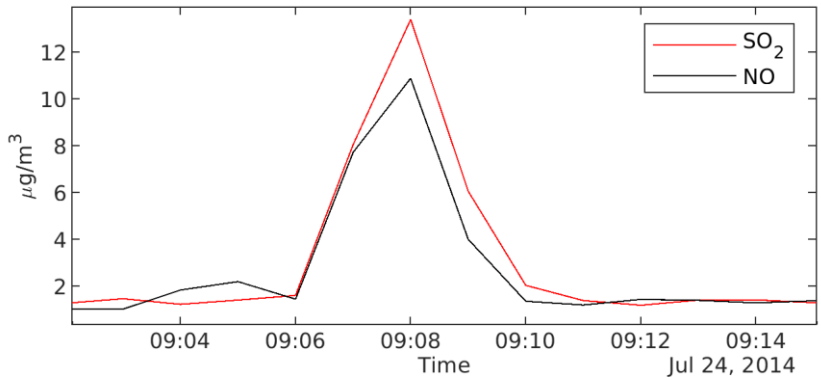

Figure 6. NO reacts rapidly with O3 and has a strong diurnal cycle and SO2:NO will too. Was CO2 not measured at the site, which would've enabled the estimation of the fuel sulfur content? If not, SO2:NOx still seems better than SO2:NO, though NOx emissions can vary significantly depending on the ship/weather conditions.

Reply: As we mentioned earlier, CO_2 measurements were not available for the whole studied period at the same location. CO_2 measurements have been available from the marine site since March 2014. Given this, we focused on analyzing the relationship between NO and SO_2. If both NO and SO_2 show concurrent peaks, it suggests a nearby emission source rather than long-distance transport, as NO would be rapidly oxidized to NO_2 over time. Therefore, we examined the SO_2:NO ratio for specific ships that operated throughout the study period from 2006 to 2020. Any significant changes in this ratio would imply shifts in fuel sulfur content, likely due to regulatory changes over the years.

Section 4.1 it's a bit odd to have this section here, when you just chosen the western sector (180-360) for the SO2:NO analysis above. Wouldn't be better to do the wind sector analysis first, and then apply the according wind sector to SO2:NO?

Reply: We thank the reviewer for his feedback. However, we respectfully disagree with this suggestion, as we chose the current structure in order to establish a logical flow and clarity of our results and discussion. Therefore, we decided to keep the structure as it is.

Figure 7. similar to figure 5, the SO2 axis is cut off at zero. Are you discarding all negative SO2 data? I don't think that is the best approach. The negative numbers (due to measurement noise) need to be kept in in order for the stats to be representative.

Reply: We would like to inform the reviewer that Figure 7 was revised to include some statistical information, as well. Moreover, we would like to clarify that none of the measured SO_2 concentrations, including any negative values due to measurement noise, were discarded during the analysis. All measurement data were applied in the creation of the figure.

However, we chose to restrict the vertical axis of Figure 7 to a range of 0-80 μg m^{-3} for better visual clarity (the range of the measurement data was from -3.5 to 178 μg m^{-3}). As a result, a negligible portion of the 1-minute measurement data, including negative values, was not shown on the figure. This decision was made for visualization purposes only and does not affect the statistical representation of the data, as the mean and median values, newly introduced in the revised figure, were calculated using the complete dataset, including negative values.

Figure 9. I don't doubt that the SECA regulation has been effective. However here the SO2 and NOx concentrations were not evaluated with consideration of plume dilution. Would've been best to normalize both gases to CO2 plume. If that's not possible, at least look at SO2:NOx ratio (which does seem to be lower after 2015).

Reply: We thank the reviewer for his suggestion, but after careful consideration and per his request, we decided to remove subsection 4.4 and thus Figure 9.

In general, I don't find that the case study has added much to the paper. Are there more information that can be teased out? E.g. fuel sulfur content before vs after 2015? Did the ship install a scrubber?

Reply: We thank the reviewer for his feedback. Therefore, after careful consideration and per his request, we decided to remove the case study part (i.e. subsection 4.4).