Answer to Referee #1 comments on "Characterization of OMI tropospheric NO2 over the Baltic Sea region" by I. Ialongo et al.

The authors thank the referee for the useful comments. Here is a point-to-point answer to the referee questions. The author text is in Roman, while the referee text is in Italic.

***1.** Focus and structure*
***a)** The manuscript contains a mixture of different aspects (the city of Helsinki, a ship track, year-to-year variations), but each of these is only touched superficially.*
According to the title, the focus of the paper is the characterization of NO2 over the Baltic Sea. To strenghten this focus, a more complete approach is needed. I.e. the discussion of cities can not be limitied to Helsinki, but has to include other cities like Stockholm and in particular Saint Petersburg. Does the lifetime/emission estimate work there as well? If not, why?

Yes we used a case study approach. To characterize the NO2 levels in this region is challenging because of the lack of data and the small emission sources. We decided to use Helsinki as case study for the urban emission and the central area of Baltic Sea to evaluate the shipping contribution, as we wanted to focus on smaller sources. The important point is that, we have now applied the Beirle's method to a relatively weak source, i.e. Helsinki area.
We will highlight modify the text as follows:

- Abstract P 2022 L10-12. This sentence will be added: "This work presents a characterization of tropospheric NO2 columns based on case-study analysis in the Baltic Sea region, using the Ozone Monitoring Instrument (OMI) tropospheric NO2 standard product."
- Introduction P2025 L5-6. This sentence will added "In particular, two case studies will be analysed: the city of Helsinki and the main shipping lane in central Baltic Sea."
- Section 2 P2026 L11. This sentence will be added: "The city of Helsinki is considered to characterize an urban site with low emissions and the central area of Baltic Sea to evaluate the contribution from ship emissions."

***b)** According to Figs. 1 and 5, there are significant NOx sources South-West of the considered region. The comparison of calm and windy conditions in Fig. 5 clearly reveals that the Baltic Sea is affected by NOx outflow from these sources. These sources have thus to be identified and their impact has to be discussed and compared to local sources.*

Yes, there are of course many land-based sources at lower latitudes (Central Europe). As explained in the text (section 2) we try as much as possible to avoid the outflow effect considering only wind speed smaller than 5 m/s and looking at central Baltic sea only. Taking into account only winds below 5m/s, the outflow from the lower left corner in Fig. 5 - central panel (the closest land-based source is located close to the south-eastern Swedish coasts) could reach the area in the black box (more than 250 km distance) only with (a daytime) lifetime about 15 h, which is not realistic for NO2. With a lifetime about 4h, which is appropriate for that latitude during summer, a wind speed larger than 15 m/s to reach the black box area. This is typically the case only for less than 3% of the pixels in the Baltic Sea area. Please note that we are concerned with daytime lifetimes of NO2. These instantaneous lifetimes hold for OMI overpass times and are usually much shorter than the 24-hours average NO2 lifetime (see e.g. Boersma et al., JGR, 2008).
Boersma, K. F., D. J. Jacob, H. J. Eskes, R. W. Pinder, J. Wang, and R. J. van der A (2008), Intercomparison of SCIAMACHY and OMI tropospheric NO2 columns: observing the diurnal evolution of chemistry and emissions from space, J. Geophys. Res., 113, D16S26, doi:10.1029/2007JD008816.

This text will be added in section 3:

- Section 3 P2030 L27. "In this case, the NO2 patterns over sea can be influenced by the air masses transported from the land sources. The closest land sources are located in southern Sweden and Denmark. With NO2 mean lifetime about 4 h, which is realistic at 55°N latitude in summer, the NO2 outflow would reach the central Baltic Sea area only with wind speed larger than 15 m/s. This is the case for only less than 3% of the grid pixels."

The different studies (ship tracks, cities etc.) should be separated by subsections.

This will be done in section 3, introducing two subparagraph for Helsinki and shipping case studies, but some aspects will remain connected to avoid repetition.

***2.** Methodology*
The applied methods are only sparsely described and there are some inconsistencies and mistakes:
***a)** OMI data*
It is stated several times in the manuscript that the OMI pixel size is 13x24 km^2. But this is only true for nadir geometry, and pixel size increases significantly towards the

swath edges. This has to be clearly stated. How are pixels at the swath edge are treated? (E.g., Beirle et al. removed the outermost 10 pixels on each side of the swath.)

Yes, the ground pixel size is at best 13×24 km in nadir and larger at swath ends. We selected only small pixels.

Section 2 P2025 L26. This sentence will be added: "Only the OMI pixel with number from 6 to 24 were included in the analysis to take into account only central small pixels and to avoid the pixels corrupted by the row anomaly."

***b)** Lifetime and emission estimate*
- The authors refer to the method proposed by Beirle et al. and indicate that they apply the same method to Helsinki. However, there are several differences with respect to the details (e.g., only 4 wind directions are considered instead of 8; a "calm threshold" of 5 m/s was chosen instead of 2 m/s etc.). Thus, a more detailed summary of the Beirle et al. approach has to be given, and different implementations have to be clearly indicated.

This text will be added in section 2:
Section 2 P2026 L3. "The methodology developed by Beirle et al. (2011) for megacities, was applied to OMI data in the Baltic Sea area, with some differences."
Section 2 P2026 L7. "…is smaller than 5 m/s (while a threshold of 2 m/s was used in the original method) analysing separately different wind directions…"
Section 2 P2026 L19. "…, while eight sectors were identified in Beirle et al. (2011)."

- In Beirle et al., emission rates are derived (mol/s), while in Ialongo et al., just a "emission factor" is given in units of molecules. What is the physical meaning of this "emission factor"? How can this E result in an NO2 line density if multiplied by an exponential decay and smoothed with a Gaussian (both unitless)? Check the units and provide emission rates instead of an "emission factor". The resulting emissions should also be compared to emission inventories.

They are actually not both unitless. The Gaussian factor has a multiplicative parameter with dimension [m^{-1}]. The factor E as reported in the equation is actually in units of molecules and that is why we specify that it should be considered as a burden parameter. We now provide the emission parameter in mol/s as E'(NO2)= E/τ=(1.5 ± 0.4) mol/s, i.e. NOx emission as NO2. We now compare also with NOx emission (as NO2) derived from EMEP database (extracted from www.ceip.at).
From EMEP database, the NOx emissions (as NO2) are E'(EMEP)=(1.8 ± 0.3) mol/s for Helsinki area (derived over the period 2007-2011), which is in agreement, within the uncertainties, with our results. The yearly emissions from EMEP database are given with uncertainty up to 15%.
This information will be reported in the manuscript as follows:

- We will add in section 2 the full equation for the fit as done in Beirle to avoid confusion.
- We will provide the emission parameters E' in mol/s and we will compare with EMEP emissions
- We will modify the text in section 3 as follows: "The resulting values for e-folding distance x0=(52±9) km, the background parameter B=(3.54±0.02)*10^22 molec./cm and the burden parameter E=(1.0±0.1)*10^28 molec. were derived from the mean fitted model (Fig. 4 - black line). The summer mean lifetime value τ=(3.0±0.5)h was then estimated by the ratio x0/w, with w=(4.9±0.2) m/s. The emission was also calculated as E'=E/τ=(1.5±0.4) mol/s. The emission parameter E' was then compared with EMEP NOx emission (given as NO2), E'_{emep}=(1.8±0.3) mol/s for the period 2007-2011 around Helsinki area, showing agreement within the uncertainties. The yearly emissions from EMEP database are given with uncertainty up to 15%. It must be noted that the emission and lifetime derived from OMI data refer to clear sky conditions. When only clear-sky pixels are considered Geddes et al. (2012), a negative bias is expected, mostly because of the accelerated photochemistry, so that both the emission E' and the lifetime would be smaller than for cloudy conditions. Despites this effect, the emission E' derived from OMI data agrees within the uncertainties with EMEP emission E'_{emep}. Furthermore, in this work a daytime NO2 lifetime is derived. This instantaneous lifetime holds for OMI overpass times and is usually shorter than the 24 h-average NO2 lifetime (see e.g. Boersma et al., 2008). "

  Boersma, K. F., D. J. Jacob, H. J. Eskes, R. W. Pinder, J. Wang, and R. J. van der A (2008), Intercomparison of SCIAMACHY and OMI tropospheric NO2 columns: observing the diurnal evolution of chemistry and emissions from space, J. Geophys. Res., 113, D16S26, doi:10.1029/2007JD008816.

- One implicit assumption of Beirle et al. is that the source is "point like" or at least symmetric (i.e. the spatial distribution can be accounted for by the convolution with a Gaussian). However, if the distribution of sources is asymmetric, this alone would cause a virtual "outflow" pattern, even without any wind, and would thus bias the fitted lifetime. This potential bias is reduced if different (in particular opposite) wind directions are fitted (as in Beirle et al.), but this is not the case here. Please discuss; does the mean line density for calm conditions look symmetric?

Yes, we checked that the line density for calm conditions is quite symmetric (you can approximately see also from fig. 3, central panel). The reason why we took into account the west wind conditions is that we have more data in that case. In some cases, when one wind direction is dominating a similar fitting can be applied as well, using only the dominating wind direction (as a function of time). (See Beirle, S., Hörmann, C., Penning de Vries, M., Dörner, S., Kern, C., and Wagner, T.: Estimating the volcanic emission rate and atmospheric lifetime of SO2 from space: a case study for Kīlauea volcano, Hawai'i, Atmos. Chem. Phys. Discuss., 13, 28695-28727, doi:10.5194/acpd-13-28695-2013, 2013.)

- The discussion of errors is very short. A simple reference to Beirle et al. is not sufficient here. If the authors claim that emissions and lifetimes can be estimated for Helsinki, they also have to provide a dedicated (and realistic) discussion of uncertainties for Helsinki, beyond the errors derived from the fit.

The text will be changed as follows:
Section 3 P2030 L6. "The errors on the estimated parameters are the standard deviations derived from the MCMC calculations. The error bars in Fig. 4 were calculated using the error propagation for the discrete integral and include the contribution from the statistical error on the mean NO2 field. The uncertainties on the emission and lifetime depend also on the error associated with OMI tropospheric NO2 column density (about 30%) and with the wind field patterns (also, about 30%). An additional uncertainty comes from the selection of the integration and the fitting intervals. In the Helsinki case, these intervals were selected to avoid the effect of high NO2 signal from the surrounding emission sources. Overall, the uncertainty on E' and τ is larger than 40%."
Please, see also the discussion on the clear sky bias in the answer to referee #2 (point 2.a)

***3.** Ship tracks*
The study of an exemplary ship track is not convincing:
***a)** The ship track seems to be interrupted at about 20.5E/58.3N. Please comment.*

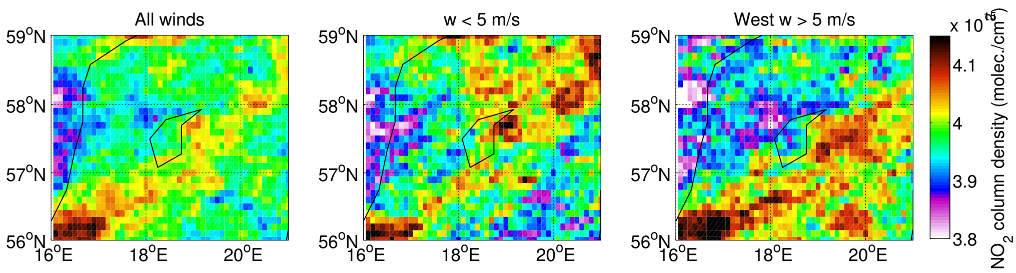
As mentioned in the paper, the signal coming from the ships is very small and comparable with the detection limit of OMI. So, despites the appropriate data screening and averaging, there are still some limitation in the dataset and the results depend on the amount of data included in the average. On the other hand, this is actually among the objectives of this work, to evaluate the sensitivity and applicability of OMI data for detecting the small ship emission signal at high latitudes. Furthermore, if we look at the ship emission data, the shipping lane tend to diverge going North, splitting in several branches. This might reduce the signal too.

***b)** The integrated NO2 amount obviously depends on the choice of the considered box. In the paper, it is close to (and downwind from!) Gotland, an Island with several oil production facilities (http://mapx.map.vgd.gov.lv/geo3/VGD_OIL_PAGE/images/Baltic_province_new_2009.jpg) and a lot of tourists during summer. Fig. 5 (a) looks like the "shiptrack" is just crossing the southern dip of Gotland. Please comment.*

This could be possible, but if we look at fig. 5 and compare with the map of the oil production locations, we should see high signal in the highest part of Gotland, where most of the oil extraction points are located and this is not the case. The black box we peaked seams to be the most representative of the marine environment including signal coming from ships. See also the answer to the next question.

***c)** Most alarmingly, the shiptrack pattern is far more distinct for windy conditions (Fig. 5)! My concern is that this may be just caused by the a-priori: The AMFs are lower over the shiptrack, as the model predicts a different (lower) profile shape, resulting in artificially enhanced tropospheric columns, as long as there is some tropospheric residue to be increased (i.e. under windy conditions, transporting NO2 from SW). This possible artefact might be ruled out by analyzing the mean (tropospheric) slant columns.*

This is how fig. 5 would look like replacing the tropospheric AMF with the geometric AMF.
The signal is still there, also under strong westerly wind conditions. So, we can exclude that this pattern is artificially produced by the AMF and OMI has truly detected NO2 signal from shipping.



Further comments:
P2023 L7: At this point, Beirle et al. is not an appropriate reference, as it neither deals with ship emissions nor the global NOx production, but focusses on Megacities.

Yes. Thank you this was a typo. We referred to Beirle et al. (2004). This will be corrected in the text.

P2024 L10: The "strong need" for monitoring NOx emissions from ships is only given if there is significant ship traffic in the Baltic sea. Please quantify.

This text will be added to the manuscript:
"Furthermore, the Baltic Sea is one of the most intensely trafficked marine areas in the world. According to IMO (2002), there are more than 2000 large vessels at any given time and about 3500–5000 different vessels are in operation in the Baltic Sea region every month."

In the reference: "International Maritime Organization: Safety Of Life At Sea (SOLAS) agreement, regulation 19, Chapter V, 1974, 2002 Amendments."

P2025 L1: I do not see the argument. As there are still high uncertainties in NOx emissions as well as chemistry, I would rather focus on strong sources at moderate latitudes, where the retrieval uncertainties are relatively low.

That is exactly the point: as there are overall still large uncertainties, even larger for small emission sources at high latitude, it is important to study tropospheric NO2. The sentence will be modified as follows:
"The uncertainties on NOx shipping and city emissions as well as lifetime estimations remain large (see e.g., Stavrakou et al., 2013), especially for relatively weak sources located at high latitudes. Thus, it is important to study the tropospheric NO2 pollution in this region."

P2029 L7-8: Why is this a "logarithmic distribution"? What I read about logarithmic distributions, they are only defined for integers and look quite different than Fig. 2.

Yes, we refer to the log-normal distribution, with longer tail in the right side. This will be corrected in the text.

P2032 L17-19: The sorting of data according to wind direction has actually been proposed and described in Beirle et al., 2011.

Yes, we wanted just to point out the potential of this approach for different applications. Perhaps the part "…, as described in this paper,…" could be misunderstood, so we'll remove that.

P2032 L20: What "good agreement with NOx emission data" is referred here?

We refer to the ship emission data from STEAM model. This will be changed as:
"The agreement with the ship NOx emission data from STEAM model confirmed that OMI NO2 data can be used to detect the signal coming from the ship emissions in a busy area like the Baltic Sea, where the effect of coastal pollution sources can mix with the emission coming from the ships to the marine boundary layer."