Answer to Referee #2 comments on “Characterization of OMI tropospheric NO2 over the Baltic Sea region” by I. Ialongo et al.

The authors thank the referee for the constructive comments. This review will certainly improve the quality of the paper.
Here is a point-to-point answer to the referee comments. The author text is in Roman, while the referee text is in Italic.

***1.** The quality of English needs to be improved. There are numerous grammatical errors, e.g, page 2024, line 25 "...being the Baltic Sea area relatively small..." —> "... being that the Baltic Sea area is relatively small..."*
Furthermore, beyond these there are many examples which sounds odd, e.g., page 2022, line 20 "...as far as they are..." —> should be "... as long as they are..."
I have pointed out a few additional examples below but there are likely several that have been missed. I suggest that once the scientific issues have been addressed that it be critically reviewed for grammar and flow by one or two English colleagues.

These mistakes have been corrected and a British colleague checked the English quality.

***2.** Analysis Details and of uncertainties*
As far as I can determine there is no analysis of uncertainty. The statistical uncertainty coming from the non-linear fitting is provided, but beyond that the only real mention is "for a complete analysis of the uncertainties see Beirle et al. (2011)". I assume that this means all other sources of uncertainty were ignored. The statistical uncertainty will be small compared to the other random and systematic sources of error, and these other sources are not even mentioned (let alone quantified). The 10% error assigned to the NO2 emission rate (E=1.0 +/- 0.1) is totally misleading. In contrast, locations analysed in Beirle et al had errors more like 50%, and these locations had larger emissions where presumably the relative errors would be smaller. Furthermore, NOx will be emitted primarily in the form on NO and not NO2.
The emissions values are only as good as their uncertainties. A detailed and convincing analysis needs to be performed for several reason, not the least of which being that there are many sceptics in this field that would not put much stock in satellite-derived emissions. Quoting uncertainties of 10% would only provide them ammunition. Beirle et al. would be a good guide for this as they have examined several sources of random error.

We introduce now the emission E’ in mol/s (as also asked by the referee n.1) and a more complete discussion of the uncertainties 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). 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%.”

Beyond that addition sources of systematic should be considered:
***a.** clear-sky bias: Only OMI measurements over clear skies are considered. How might this bias the results? There are a couple of papers that have looked at this: Geddes et al. (Remote Sensing of Environment, 2013), McLinden et al. (ACPD, 2014).*

This aspect could have a role when comparing the emission and lifetime estimate with existing database (as we do now reporting the emission in mol/s and comparing to EMEP data (available at www.ceip.at)).
According to the literature there two ways the clear sky bias would be produced. (I think the correct reference was Geddes et al. (2012)).

First, there is the effect of wind patterns. Removing cloudy data would result in taking into account only certain wind directions/patterns. In our case, the wind patterns for clear sky conditions are very similar to the one obtained under all cloud conditions. Please see the figure at the bottom of this file, where the wind patterns in Helsinki and central Baltic Sea areas for clear sky pixels are shown. If you compare this picture to Fig.2 in the original manuscript, where all cloud conditions were considered, you can notice that there is basically no difference between them. Furthermore, when calculating the emission and lifetime, we already consider only winds from East to West, as the methodology is applied under specific wind conditions. Thus, we do not expect to see a strong effect of wind patterns due to the clear sky screening.
Second, there is the effect of accelerated photochemistry. In particular a shorter lifetime and a higher NO2 photolysis rate are expected under clear sky conditions. There is no possibility to sample the yearly emission data used for comparison according to OMI clear sky criteria but this potential effect will be discussed in the paper as follows:

“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. Despite this effect, the emission E' derived from OMI data agrees within the uncertainties with EMEP emission E'_{emep}.”

Geddes, J.A., Murphy, J.G., Celarier, E.A., and O'Brien, J.: Biases in long-term NO2 averages inferred from satellite observations due to cloud selection criteria, Remote Sensing of Environment, 124, 210-216, 2012.

b. *GMI model used in SP retrieval: The emissions used by the GMI model are from 1997 or 1998, and these impact the profile shapes and thus air mass factors and VCDs. How have emissions in the Baltic area changed since then and discuss how this could bias your emissions numbers.*

We use the SP Version 2, where the monthly mean NO2 profile shapes derived from GMI CTM multiannual (2005–2007) simulation, are used (see Bucsela et al., 2013). We also evaluated the effect of a-priori profile on AMF over sea under strong wind conditions to answer to a question from referee #1. Replacing the tropospheric AMF with a geometrical AMF, we can observe than the signal coming from the ships is still present in the Baltic Sea area. So, the AMF does not produce artificial signal over sea, under strong outflow situations, but it most probably comes from ships (Please see the picture in the answer to referee n.1).

c. *Winds: What ECMWF reanalysis was used, what is its resolution. Why use winds at 950 hPa? This corresponds to what, 250 or 300 m? I am guessing that at the locations considered here the wind speed increases rapidly with altitude and so an average wind speed over the boundary layer could be twice what are used here. This would have large implications on the derived lifetime and emission rate. Discuss this.*

We use the average below 950 hPa as in the original method (1000, 975 and 950 hPa), to account for the quick vertical mixing. We also evaluated the differences in NO2 pattern using these 3 levels separately and the mean patterns were very similar. Overall, the uncertainty related to effect of the wind fields is in the order of 30%. The spatial resolution is 0.25 degrees. This information will be added in the manuscript.

Other comments:
page 2022, line 7: Measurements over snow will help with signals. Rework this sentence to as to not mix up the two issues (snow –> high signals, other complicating issues / high latitude –> lower signals)

The sentence will be changed as: “Tropospheric NO2 monitoring at high latitudes using satellite data is challenging because of the reduced light hours in winter and the small signal due to low Sun, which make the retrieval complex.”

page 2022, line 16: Provide the emission rate in terms of mass as this is more useful, either in addition to molecules or instead of. Is this an annual amount or for the summer only. It should be converted into a rate.

As mentioned before we introduce now the emission parameter E’ in mol/s as in the original paper and in order to be compared with EMEP emission data (please, see answer to question n.2).

page 2024, line 28: one exception is the oil sands work of McLinden et al (GRL 2012), you should add this as a counter example

Yes, good point. This reference will be added to the manuscript as:
“One exception is the paper by McLinden et al. (2012), who looked at the air quality over the Canadian oil sands using satellite data.”
McLinden, C. A., V. Fioletov, K. F. Boersma, N. Krotkov, C. E. Sioris, J. P. Veefkind, and K. Yang (2012), Air quality over the Canadian oil sands: A first assessment using satellite observations, Geophys. Res. Lett., 39, L04804, doi:10.1029/2011GL050273.

page 2023, line 6: "as they represent a relevant part" ... I assume you mean the portion *of ship emissions is large enough that they need to be considered. "Relevant" means connected with or pertinent. I would suggest you rephrase this using "as they represent a sizeable fraction" or something similar*

Corrected

page 2024, line 7: "could increase" - this sounds odd since 2012 is in the past. use "may have increased" or something analogous.

Corrected

page 2025, line 1: "remain still large" —> "remain large"; likewise "lifetime estimations" is probably better phrased as "estimates of lifetime"

Corrected

page 2026, line 1: Why consider only June-August? Why not May and September? These should also be snow free and will increase you signal to noise.

We analysed these months too, but the data were extremely unevenly distributed, with many missing data over the area of interest. We wanted to avoid the situation where only a few overlapping pixels (or just one pixel so, no overlapping pixels at all) would have determined the monthly mean. That's why we limited the analysis to summer months. The use of spring and autumn months needs a more careful analysis, which could be topic for future work.

page 2026, line 5: The local time of the Baltic is UTC+2? State this here so that readers know that UTC 12:00 is a good match with the OMI overpass time. It is not obvious otherwise.

This sentence will be added to the manuscript: "The local time in the area of interest ranges between UTC+1 and UTC+2, which approximately corresponds to the nominal overpass time of OMI (13:45 LT)."

Figure 1: It is difficult to make out the letters in the panels on the left.

The letters are now in a different position, so that there is no overlap with the coastlines. Hopefully that is clear enough.

Figure 2: Figure 2 shows the distribution of winds. Have these been sampled in the same way as OMI (considering only clear skies)? If not, they should be as this can dramatically change the patterns. Redo these, or add additional panels, showing the clear-sky wind distributions.

We follow your recommendation replacing Fig.2 with the distribution of winds sampled as OMI. The patterns do not change much, though (see the picture below in comparison with Fig. 2 in the original manuscript).