I copied here the answer to the referees' comments, which are already published in the public discussion.

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 km². 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 lalongo 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 r=(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/r=(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%."

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."

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 $\tau = (3.0 \pm 0.5)$ h was then estimated by the ratio x0/w, with w=(4.9\pm0.2) m/s. The emission was also calculated as $E'=E/\tau=(1.5\pm0.4)$ mol/s. The emission parameter E' was then compared with EMEP NOx emission (given as NO2), E'empe=(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 <u>www.ceip.at</u>)). 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).

