

The reply to the anonymous referee #1 (RC1)

We are grateful to the referee for the very insightful comments. We took them into account while preparing the revised version of the manuscript.

Below, the actual comments of the referee are given in **bold courier font and blue colour**. The text added to the revised version of the manuscript is marked by **red colour**.

1) **The abstract presents a lot of technical details, such as the data processing activities in four steps. I recommend to remove these.**

The text about four steps of data processing has been removed from the abstract.

2) **Part of the methodology is based on emission assessments using differential column measurements equipped with two solar-tracking spectrometers upwind and downwind of the city. The authors could consider to include Chen et al. (2016): "Differential column measurements using compact solar-tracking spectrometers", where the same principle has been used, as a reference in line 100.**

We added the suggested reference in several places, in particular:

Chen et al. (2016) developed and used differential column methodology (downwind-minus-upwind column differences) for the evaluation of CH₄ emissions from dairy farms in the Chino area.

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The idea and the methodology of EMME experiment were based mainly on the studies by Hase et al. (2015), Ionov and Poberovskii (2015), Chen et al. (2016) and Viatte et al. (2017).

3) **Page 9: The authors have determined the optimum integration time by examining the "half width" of the short term variations. Another possibility to determine the optimum integration time is to use the Allan variance analysis. This approach was used in Chen et al. (2016).**

We added the following text at the end of Section 4.2:

The chosen averaging interval of 15 min is in good agreement with the estimation of the optimal integration time (10 min) obtained as a result of the Allan analysis implemented by Chen et al. (2016). Chen et al. (2016) applied this approach to the differential measurements of XCO₂, XCH₄ performed by three EM27/SUN spectrometers within urban areas.

4) **Page 9: please add units to the parameters denoted in equation (1).**

In the revised version units are added to the parameters denoted in equations (1-3).

5) **Section 4.4: I have doubts about the definition of the effective air parcel path length. By deriving the effective path length including only the "polluted path", and excluding the "clean path", you are determining the emission flux of the industrial and traffic (the polluted areas), but not the emission flux of the whole city. So it could be not fair to compare these numbers to the emission inventories of the city, which may result in much higher emissions compared to the emission inventory.**

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12) Table 4: The big discrepancies between the estimate in the paper and the emission inventory could be partially attributed to the usage of the effective path length, so the flux density determined in this study is focused on the industry area and traffics whereas the inventory is the averaged flux in the city. Please discuss this possibility.

The main goal of the field campaign is to evaluate the area fluxes (F) originated from the urbanized territories of the St.Petersburg agglomeration. Therefore we excluded from the consideration the territories of parks, forests and water bodies as the areas that practically have no anthropogenic emission sources. At the same time we agree with the referee’s statement that “So it could be not fair to compare these numbers to the emission inventories of the city, which may result in much higher emissions compared to the emission inventory”. In the revised version of the manuscript, we estimated the urbanized area of the St.Petersburg agglomeration according to the land-use classification that was developed for the derivation of the effective path lengths. We obtained that the total urbanized area of the agglomeration occupies about 984 km² while the official area of the entire St.Petersburg is 1439 km². Therefore the values of area fluxes for all gases (CO₂, CH₄, CO and NO₂) that were estimated using the official inventory data have been recalculated and, as a result, became higher. Revised version of Table 1 (the former Table 4) is given below. The changes are highlighted by yellow colour.

Table 1. Area fluxes for CO₂ (kt km⁻² yr⁻¹), CH₄ (t km⁻² yr⁻¹), CO (t km⁻² yr⁻¹) and NO_x (t km⁻² yr⁻¹) obtained during EMME-2019 and the flux estimates for St. Petersburg based on in situ measurements. The values previously reported in literature are also presented.

Area flux	EMME		In situ measurements	Literature sources	
	(9 days)	(4 days)		St. Petersburg	The world’s cities
1	2	3	4	5	6
CO ₂ , kt km ⁻² yr ⁻¹	89 ± 28	85 ± 12	40 ± 30	31 (Serebritsky, 2018), 46 (EDGAR database, 2018) 6 (suburbs, Makarova, 2018)	29 (London, O’Shea, 2014) 35.5 (London, Helfter, 2011) 12.8 (Mexico City, Velasco, 2005) 12.3 (Tokyo, Moriwaki and Kanda, 2004) 0.8 – 7.7 (Krakow, Zimnoch, 2010) 28.3 (Berlin, Hase, 2015)
CH ₄ , t km ⁻² yr ⁻¹	135 ± 68	178 ± 30	120 ± 80	25 (Serebritsky, 2018, 2019), 110 (Makarova, 2006), 44 (suburbs, Makarova, 2018) 32 (suburbs, Zinchenko, 2002)	66 (London, O’Shea, 2014) 7 – 28 (Krakow, Zimnoch, 2010)
CO, t km ⁻² yr ⁻¹	251 ± 104	333 ± 103	90 ± 50	410 (Serebritsky, 2018, 2019), 390 (Makarova, 2011), 90 (suburbs, Makarova, 2018)	106 (London, O’Shea, 2014) 1520 (Mexico City, Stremme, 2013)

NO _x , t km ⁻² yr ⁻¹	66 ± 28	-	-	69 (Serebriksy, 2018, 2019)	63-252 (London, Lee, 2015) 13- 300 (Norfolk, Marr, 2013)
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We see that even in this case the official inventory data provide much lower area fluxes for CO₂ and CH₄. The validity of our results can be confirmed if we consider the values of emission ratio (ER) which are widely used as a characteristic of the relative structure of emissions from a source. If we compare ERs estimated from our observational data (FTIR measurements during EMME campaign and in-situ routine observations of CO₂, CO and CH₄) and ERs derived from official inventory data, we can see that these values differ significantly from each other, see Table 2 (the former Table 5) in the paper. For example, the mean value of ER_{CO/CO2} obtained from our observations varies from 5.9 to 6.2, at the same time the ER_{CO/CO2} value estimated using official inventory data equals to 21. This difference in ER_{CO/CO2} values obtained using “top-down” and “bottom-up” approaches could be explained by the underestimation of total CO₂ and CH₄ emission of St.Petersburg in the official inventory.

6) Line 358: repetition of “April 25”, please delete the second one.

Repetition of “April 25” has been deleted.

7) Equation 2: It is not clear what kind of wind speeds are taken for the consideration, please elaborate it.

We added the following text:

... where δV is the relative variation of the wind speed over a day estimated using HYSPLIT meteorological data,...

8) Equation 2: you can determine the square root of the error terms instead of adding them

The esteemed referee is perfectly right. The assumption of uncorrelated errors of input parameters should work well in our case. However, in order to be on the safe side we decided to present the estimation of the upper limit of the total error (completely correlated errors of wind and TC which are anticorrelated with the errors of L), therefore we added terms instead of using the square root of the sum of squared terms. In the original version of the manuscript we have already written: “The δF values calculated in this way can be considered as an upper limit of the F uncertainty.”

9) Figure 5: there is no unit for the color bar [0-25]. The river is drawn as blue, but it looks confusing because the blue color is also assigned to the color bar.

In the revised version we changed the figure caption (Fig.3, former Fig.5):

The HYSPLIT model output for each of the campaign days (10:00 UTC) used as the forecast of the megacity plume while planning the field campaign. The colour bar units for TC_{NO2} are [0-25] 10¹⁵ cm⁻². The blue line in the southeast indicates the river Neva.

10) Figure 7: you could show the scaled results instead. It will illustrate how the close the curves are to each other after the scaling process.

Figure 7 (at present Fig. 5) in the original manuscript is showing the data after the scaling process. However, it was not indicated explicitly. In the revised version we give this information in the text of the article and in the figure caption:

The scaled results of the side-by-side measurements of XCO₂, XCH₄, and XCO by FTS#80 and FTS#84 on 12 April 2019 at the St. Petersburg observational site are presented in Fig. 5.

Figure 5: The scaled results of the side-by-side measurements of XCO₂, XCH₄, and XCO by FTS#80 and FTS#84 on 12 April 2019.

11) **Figure 8: It is not very clear from the description which paths you took for determining the effective path length, are these paths from different days? Please elaborate these further. Do you have only one effective path length for all the days for each meteorological data set (LOCAL, GDAS, and HYSPLIT)? If so, how the effective path lengths vary given by different meteorological data set?**

Figure 6 (former Fig.8) shows all the paths of our experiments, one path per day. They are all different, since the FTIR observation locations and the wind field change from day to day. In the original manuscript we announced in the figure caption that **for simplicity, the path lengths on the map are equal**. We agree that this phrase can be misleading. So, in the revised version the figure caption is changed:

“An example of linear backward paths (black straight lines, black dots show the downwind FTS locations) for the days of FTIR observations. The major land use classes are shown by different colours (blue for the water bodies, grey for the residential buildings/industrial areas, green for the parks and forests). The path lengths on the map are plotted equal only for illustrative purpose. In fact they are all different since the FTIR observation locations and the wind field change from day to day. Red line designates the official administrative boundary of the St. Petersburg agglomeration. Red "star" indicates the location of one of the major thermal power stations (TPS) located to the north of St. Petersburg. Map data © 2019 Yandex.”

Special notes:

A number of typos have been found and corrected during the preparation of the revised version of the manuscript. All of them are not critical with respect to the results and conclusions.

We slightly rearranged the text by moving several small parts of the text to other places without any changes. The general structure of the article remained unchanged. This minor rearrangement was a result of revising the manuscript in accordance with the comments and suggestions of referees.

Maria Makarova
on behalf of all co-authors