Authors' reply to Dr. Timofeyev

We are grateful to Dr. Timofeyev for his interest in our paper.

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.

***The problem investigated in the study is important and relevant due to the Earth climate change and the importance of megacities for the variation of the atmospheric gas composition. Therefore, the authors should be welcomed to keep providing studies on the independent assessment of such emissions.**

We thank Dr. Timofeyev for this positive assessment of our work.

***The different estimates of the St.Petersburg integral emissions which are in range from 44800 to 74800 kt/year are given in the article. The difference between the minimum and maximum of the emissions constitutes approximately 31000 kt/year or ~70% relatively to the minimal value. The variations have to be analyzed, the inaccuracies of the approaches applied and natural variations have to be assessed. What is the reason for such a big spread between emission estimates - the technique of the measurements, lack of the observation data or their quality, the natural emission variation, the influence of the different trajectories, etc? The analysis of the estimated emissions and their uncertainties (random and systematic), the measurement technique and the inversion modelling approach used in the study have to be provided in the article.**

Actually, our study reveals 75800±5400 kt/year from the field measurements in 2019 and 68400±7100 kt/year from the field measurements in 2020. Thus the difference between these two is just 10%, so the "70% difference" is definitely out of the question here. Apparently, the 10% difference between the two estimates in 2019 and 2020 is rather small and looks quite reasonable. As for the value of 44800 kt/year – this is an estimate based on the analysis of the ground-level measurements of CO_2 surface concentrations, carried out by gas analyzer at the site of Peterhof (see the subsection 3.3 "Simulations of ground-level CO_2 concentrations" of the original manuscript for full details). This estimate stands aside, and the reasons for this are discussed in the original version of our manuscript (subsection 4.1 "The results of the EMME-2019 campaign"):

> *Resulting CO_2 emission rate is almost twice as high as the above estimate, based on the analysis of ground-level CO_2 measurement data (Section 3.3, 44800±1900 kt year^{-1}). This difference may indicate a significant contribution of elevated CO_2 sources (industrial chimneys) that could not be registered by the ground-level in situ measurements, as the elevated exhausts of pollution are more likely to further rise up, rather than descend to the ground. In contrast, FTIR measurements of the total column keep being sensitive to this kind of emissions. In addition, while FTIR measurements implement a "cross section" of the urban pollution emission zone in a series of multidirectional trajectories (depending on the wind direction), local ground-level in situ measurements at a specific location (Peterhof) can not capture the contribution of the entire mass of urban emissions. Thus, estimates of integral CO_2 emissions based on the interpretation of ground-level measurements in Peterhof can be considered as a lower limit of an estimate.*

***The significant systematic errors of the integral emission estimation approach used in the study can be related to the trajectories applied in the approach. The analysis of the Fig.6 demonstrates that the trajectories which link the positions of the observations cover the city irregularly. For instance, there are large city`s areas which were not covered by the trajectories completely. By contrast,**

some of the city's zones were covered by the measurements (which after that were used in the emission estimation) several times.

First, we would like to emphasize that there are no "significant systematic errors of the integral emission estimation approach" reported in our paper (see the comment above). On the contrary, the differences in estimates are quite insignificant. Second, the air mass trajectories shown in Fig.6 relate to the problem of determining the area fluxes by the mass balance approach adopted in the form of a one-box model. This part of the paper is a very small element of our study, and it is intended to demonstrate the agreement with similar results presented earlier by Makarova et al. (2021). This section has nothing to do with the main task of determining the integral CO_2 emission based on the comparison of FTIR measurements with HYSPLIT simulation data.

***Since the quality of a priori information (especially the accuracy of a transport model) is crucial for the quality of inverse modelling, readers can be interested by the comparison of the local measurements of CO_2 mixing ratio in Peterhof and HYSPLIT modelled data. The quantitative analysis (STD, MAE, RMSE) of such comparison before and after the scaling of the a priori emissions have to be provided in the study.**

This is exactly what we did and what is already available in the original version of the manuscript. We have the impression that Dr. Timofeyev missed section 3.3 "Simulations of ground-level CO_2 concentrations":

> *Routine measurements of CO_2 surface concentrations have been carried out at the atmospheric monitoring station of St. Petersburg University in Peterhof (59.88° N, 29.82° E) since 2013. These observations are the in situ measurements using a gas analyzer Los Gatos Research GGA 24r-EP. The instrument is installed on the outskirts of a small town of Peterhof in the suburbs of St. Petersburg (see location in Fig. 1). This place is far enough away from busy streets and other local sources of pollution, with an ambient air intake being 3 meters above the surface. To test the HYSPLIT model setup for the St. Petersburg region, we calculated the surface concentration of CO_2 near the Peterhof during the 2019 EMME measurement campaign – from March 20 to April 30, 2019 (Makarova et al., 2021). The results of the model calculations were compared to the data of in situ measurements (due to the instrument failure in 2020 the comparison is limited to the period of EMME campaign in 2019 only). Observational data and simulation results were averaged over 3-hour intervals. The resulting comparison is shown in Fig. 4. The model reproduces the temporal variations of CO_2 including the main periods of significant growth of concentration; the correlation coefficient between the calculation and measurements is equal to 0.72. The background value of the surface concentration is taken as 415 ppmv based on long-term local measurements. It is important to emphasize that quantitative agreement is achieved by linear scaling of the a priori integral urban CO_2 emission. The scaling coefficient for emissions corresponds to the value of the integral urban CO_2 emission from the territory of St. Petersburg of 44800±1900 kt year^{-1} (the given uncertainty is due to the uncertainty of the fitted scaling factor). This value is noticeably higher than official estimates mentioned above and ODIAC data for 2018 (32529 kt). The average discrepancy between the measurement and simulation data shown in Fig. 4 is 2±9 ppmv (model calculations are systematically lower).*

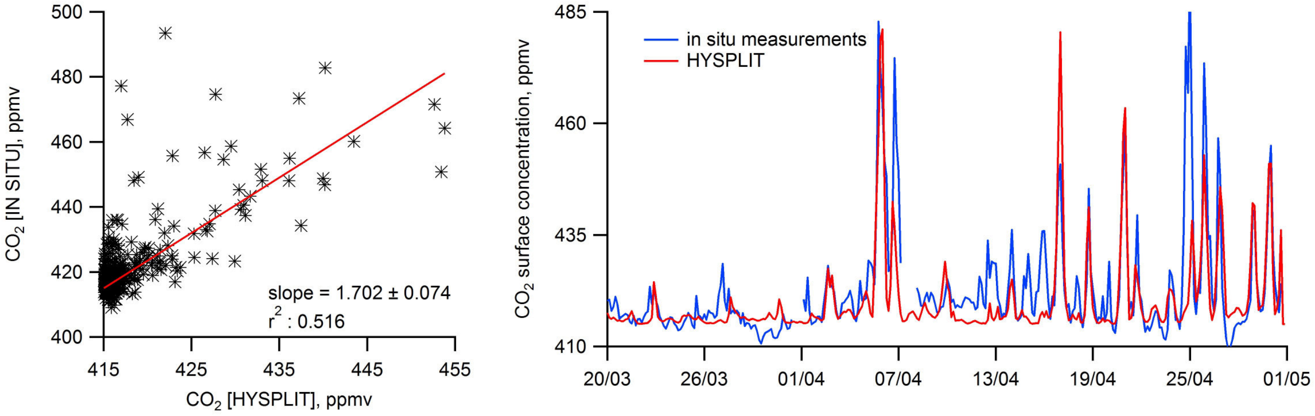

Figure 4: Comparison of the HYSPLIT simulations and the in situ measurements of surface CO_2 concentration in Peterhof (59.88° N, 29.82° E) in March-April 2019. Left panel: The values of surface CO_2 compared with the results of HYSPLIT simulations before scaling of the ODIAC emissions data. Right panel: HYSPLIT data obtained using scaled ODIAC CO_2 emissions compared with observed surface CO_2. Measurement and simulation data are averaged over 3-hour intervals.

***The authors give insufficient review on the CO_2 and other greenhouse gases emission estimates provided for Moscow and St.Petersburg megacities by other researchers.**

To our knowledge, there are no other studies of the CO_2 emissions either in St. Petersburg or in Moscow megacities published in the peer-reviewed scientific literature and relevant to the specific topics of our research. Dr. Timofeyev in the beginning of his comment mentions the paper by Y.M. Timofeyev, G.M. Nerobelov, Y.A. Virolainen, A.V. Poberovskii, and S.C. Foka: "Estimates of CO_2 anthropogenic emission from the megacity St. Petersburg", Dokl. Earth Sc. 494, 753-756 (2020), https://doi.org/10.1134/S1028334X20090184. We are certainly aware of this work, since the two of its co-authors – Y.A. Virolainen and S.C. Foka – are active participants of both 2019 and 2020 EMME campaigns and they are the co-authors of our present paper. The mentioned study by Dr. Timofeyev et al. (2020) basically exploits the experience of EMME-2019 campaign and confirms one of the main of its findings – the twofold underestimation by the official inventory of the CO_2 emission. This finding had been already reported in the discussion paper submitted by Makarova et al. to AMT in April 2020 and later on accepted for publication and published in the beginning of 2021. Dr. Timofeyev et al. in their work used the data of observations performed within EMME-2019 and combined it with an ODIAC emissions inventory. It should be emphasized that according to the tradition of the Russian Academy of Sciences articles are submitted to the Journal “Dokl. Earth Sc.” only with a recommendation from a member or corresponding member of the Academy and these articles do not go through a standard peer-review process (the article by Timofeyev et al. (2020) was received June 16, 2020; revised June 17, 2020; accepted June 18, 2020). To our opinion, the absence of any peer review has led to the fact that this paper may contain inaccuracies. For example, a reader can come to the conclusion that the authors used an extremely simplified assumption for the transfer of air masses: strictly straight from the measurement point on the upwind side to the measurement point on the downwind side (see Fig. 1 on page 754, Timofeyev et al., 2020). If so, then this approach is rather questionable, since in reality the wind direction has never exactly coincided with the straight line connecting these two measurement points. We would like to emphasize that the article by Timofeyev et al. (2020) is extremely short and is missing a lot of information which is important. For example, the authors mention the tomographic approach to the analysis of measurement data by, but do not reveal the essence of this method at all. It should be noted that the authors did not acknowledge the owners of the used equipment (FTIR spectrometers EM27/SUN from Karlsruhe Institute of Technology, Germany) and the contribution of the German participants of the EMME experiment. Timofeyev et al. (2020) did not specify the sources of funding for the measurements campaign (European Union's Horizon 2020 research and innovation programme under grant agreement No 776810, VERIFY project; Russian Foundation for Basic Research through the project No.18-05-00011). To the first glance, the absence of this information may seem a formal fault, however, to our opinion, it can mislead readers and produce a wrong impression about the EMME experiment and the priorities of the obtained results. Therefore, we decided to avoid citing the mentioned article by Timofeyev et al. (2020) in our present paper.

***A descriptive table containing details of the 2020 measurement campaign (e.g. atmospheric conditions with its dynamic, etc) has to be added to the article how it was done in the previous study.**

We supplied Section 2 "Methods and instrumentation" of the revised manuscript with the tables of information about each mobile experiment in 2019 and 2020 (see below):

> Details of both field campaigns are given in Tables 1 and 2 for 2019 and 2020, respectively. The tables contain the Fourier transform spectrometer (FTS) instrument IDs (#80 and #84 in 2019, #84 in 2020), the position on the upwind and downwind sides of the city (latitude and longitude), and the duration of observations. Note that each experiment presented in the tables consists of a pair of series of measurements – from the upwind and downwind sides. In 2019, observations of two FTS instruments (#80 and #84) simultaneously were used for this purpose (see Table 1). In 2020 the single FTS instrument (#84) was moved between the upwind and downwind positions (see Table 2). The average duration of measurements in 2019 was 3 hours within the period of ~12:00-15:00. In 2020, the duration of the measurements was limited to about 1 hour (sometimes less), and the observation time varied from 11:00 to 19:00. Since a single instrument was used in 2020, the time difference between upwind and downwind measurements in 2020 ranged from 3 to 5 hours.

Table 1. EMME-2019 field campaign details: the dates of experiments in 2019 and the locations of FTS instruments during the upwind and downwind observations. The data on the direction and speed of the surface wind correspond to observations at one of the meteorological stations in the center of St. Petersburg at local noon (http://rp5.ru/Weather_archive_in_Saint_Petersburg, last access: 11 March 2021).

No.	Date	Wind speed, ms^{-1}	Wind direction	FTS identifier (instrument #) location (latitude, longitude) observation time (local)	
				upwind	downwind
1.	21 March 2019	3	WSW	#80 59.88°N, 29.83°E 14:07-15:07	#84 59.95°N, 30.59°E 13:08-15:36
2.	27 March 2019	2	WSW	#84 60.01°N, 29.69°E 11:49-15:08	#80 59.85°N, 30.54°E 11:42-14:57
3.	01 April 2019	3	WSW	#84 60.01°N, 29.69°E 11:01-13:24	#80 59.85°N, 30.54°E 11:15-14:31

4.	03 April 2019	3	S	#84 59.88°N, 29.83°E 14:47-16:02	#80 60.04°N, 30.47°E 11:57-14:21
5.	04 April 2019	3	SW	#84 59.81°N, 30.09°E 11:59-14:16	#80 60.04°N, 30.47°E 11:59-14:16
6.	06 April 2019	2	SE	no.84 59.95°N, 30.59°E 12:14-15:23	no.80 60.01°N, 29.69°E 12:15-15:29
7.	16 April 2019	2	NE	#84 60.01°N, 29.69°E 11:13-15:08	#80 59.86°N, 30.11°E 11:21-14:59
8.	18 April 2019	2	NE	#80 60.04°N, 30.47°E 12:07-14:56	#84 59.81°N, 30.09°E 11:38-15:24
9.	24 April 2019	1	WSW	#84 60.01°N, 29.69°E 11:38-14:55	#80 59.85°N, 30.54°E 11:52-15:22
10.	25 April 2019	1	WSW	#80 60.04°N, 30.47°E 12:07-14:49	#84 59.81°N, 30.09°E 11:19-15:08
11.	30 April 2019	2	SSE	#80 59.85°N, 30.54°E 12:35-13:31	#84 60.01°N, 29.69°E 12:22-13:46

Table 2. EMME-2020 field campaign details: the dates of experiments in 2020 and the locations of FTS instrument during the upwind and downwind observations. The data on the direction and speed of the surface wind correspond to observations at one of the meteorological stations in the center of St. Petersburg at local noon (http://rp5.ru/Weather_archive_in_Saint_Petersburg, last access: 11 March 2021).

No.	Date	Wind speed, ms^{-1}	Wind direction	FTS identifier (instrument no.) location (latitude, longitude) observation time (local)	
				upwind	downwind
1.	22 March 2020	1	N	#84 60.11°N, 30.48°E 10:38-11:55	#84 59.94°N, 30.40°E 13:17-14:38
2.	22 March 2020	1	N	#84 60.11°N, 30.48°E 10:38-11:55	#84 59.81°N, 30.14°E 15:55-17:16
3.	23 March 2020	2	W	#84 59.93°N, 29.64°E 12:55-14:33	#84 59.90°N, 30.52°E 16:24-18:02
4.	27 March 2020	2	WSW	#84 59.88°N, 29.83°E 10:35-11:51	#84 59.94°N, 30.60°E 13:24-14:12
5.	27 March 2020	2	WSW	#84 59.88°N, 29.83°E 10:35-11:51	#84 59.96°N, 30.60°E 14:34-15:15
6.	05 April 2020	4	WSW	#84 59.82°N, 29.96°E	#84 59.83°N, 30.52°E

				12:44-13:43	10:53-11:48
7.	08 April 2020	3	WSW	#84 59.89°N, 29.89°E 14:58-16:46	#84 59.83°N, 30.52°E 11:09-13:43
8.	01 May 2020	1	ESE	#84 59.73°N, 30.25°E 18:01-19:03	#84 60.05°N, 30.06°E 13:22-14:27
9.	01 May 2020	1	ESE	#84 59.73°N, 30.25°E 18:01-19:03	#84 60.03°N, 30.00°E 15:10-16:11