Authors' reply to referee #1

We express our gratitude to the referee for reviewing our paper and for the valuable corrections and suggestions to improve our work. As an outcome, we have carefully revised the manuscript, with a number of modifications being done in compliance with all the raised issues.

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.

`*General structural comments:`

`*1. There are no clear distinction between the methods, results and discussion sections in the manuscript. The manuscript should be reorganized in order to better guide the audience through the entire analysis step by step.`

In response to the referee's comment, we restructured our article as follows:

1 Introduction
2 Methods and instrumentation
2.1 Bruker EM27/SUN FTS and spectra processing
2.2 A priori data on FF CO_2 emissions
2.3 The HYSPLIT model general setup
2.4 Test simulations of ground-level CO_2 concentrations
3 Evaluation of integrated CO_2 emissions from field FTIR measurements
3.1 The results of the EMME-2019 campaign
3.2 The results of EMME-2020 and comparison with EMME-2019
4 Summary and conclusions

`*2. The introduction doesn't clearly outline the scope of the work. The bullet points at the end need to be expanded in more details. All the subsequent sections and subsections that follow the introduction should be outlined in the introduction in the same order.`

In response to the referee's comment, we have rewritten this of introduction as follows:

The present study continues the analysis of the data of EMME-2019 and demonstrates the first results of the 2020 campaign. We concentrate our efforts on the CO_2 emissions leaving the results relevant to other gases beyond the scope of the research. As an extension to the work by Makarova et al. (2021) our goal in this paper is to estimate the integral CO_2 emission by St. Petersburg megacity rather than area fluxes. Completing this task consists of the following basic steps:
- We use mobile FTIR measurements to obtain CO_2 column enhancements (ΔCO_2) related to urban anthropogenic emissions.
- We adapt the ODIAC database (Oda and Maksyutov, 2011) to construct a priori information on the spatio-temporal distribution of anthropogenic CO_2 emissions on the territory of St. Petersburg.
- We initialize the HYSPLIT dispersion model, HYbrid Single-Particle Lagrangian Integrated Trajectories (Draxler and Hess, 1998; Stein et al., 2015) with the ODIAC emissions to simulate CO_2 3D field over the city of St. Petersburg.
- We evaluate the performance of our HYSPLIT model setup by calculating the surface CO_2

concentrations and comparing them with the routine in-situ measurement results (Foka et al., 2019).
- We scale the emission input data for the HYSPLIT model simulations in order to reproduce the observed ΔCO_2.
- Finally, from the scaled emission a priori data we get the estimate of integral CO_2 emission by St.Petersburg.

The paper is organized as follows. Section 2 describes the methods and instruments, including a description of the EMME measurement campaign and the equipment used, methods for processing the measurement results, the configuration of the HYSPLIT model and its evaluation based on calculations of ground-level CO_2 concentrations. Section 3 presents main results of EMME-2019 and EMME-2020 including estimates of integrated CO_2 emissions derived from FTIR measurements of the 2019 and 2020 field campaigns, combined with HYSPLIT model simulations. Section 4 contains a summary of our findings.

*3. The authors should not assume that the readers are familiar with the previous work neither they should expect them to read other manuscripts to understand the scope or the methods used in the current study. While citation to the earlier study (Makarova et al. (2020)) is encouraged in moderation, methods should be briefly explained in the current manuscript as well.

We have added more information about the 2019 and 2020 field measurements to the new Section 2 "Methods and instrumentation".

*Scientific comments:

*1. It is mentioned that during the 2019 campaign two FTIR instruments were used whereas only one instrument was deployed during the 2020 campaign. There are two questions regarding this: 1) Where were the locations of the two instruments in 2019? If there's more than one configuration, clearly outline the locations at each specific day. Presenting the measurement locations on the map is encouraged. 2) How did you estimate ΔCO2 in 2020 by using only one instrument. Did you move the instrument within one day? If yes, what time? Outline clearly maybe in a table.

The strategy of our mobile experiments was to conduct paired series of measurements – at the upwind and downwind sides of the city. This was noted in the text of the article at the beginning of Section 2 in the original manuscript version (*"The EMME measurement campaign (short summary)"*): "*two mobile EM27/SUN FTIR spectrometers were used in the field experiment for simultaneous measurements inside and outside of the air pollution plume*" (in 2019) and "*one spectrometer which was transported between clean and polluted locations within one day*" (in 2020). Following the reviewer's comments, we expanded this part of the text and included the tables with information about each mobile experiment in 2019 and 2020 (see below, beginning of a new Section 2 in the revised manuscript, "Methods and instrumentation"):

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 12:44-13:43	#84 59.83°N, 30.52°E 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

In addition, detailed information about the location of the instruments for the selected days is presented in separate figures provided in the Appendix A:

Appendix A: Location of ground-based measurement points with respect to the urban pollution plume.
The location of FTS field measurements is shown on the maps of vertically integrated CO_2 (total column: TC) produced by HYSPLIT for selected campaign days in 2019 and 2020 (10:00 UTC), see Fig. A1 and A2. The locations of the FTS instruments on the upwind and downwind sides are indicated by blue and red circles, respectively. Note that in 2020 there were days when the downwind measurements were performed twice, at different locations – on March 23 and May 1 (see Fig. A2).
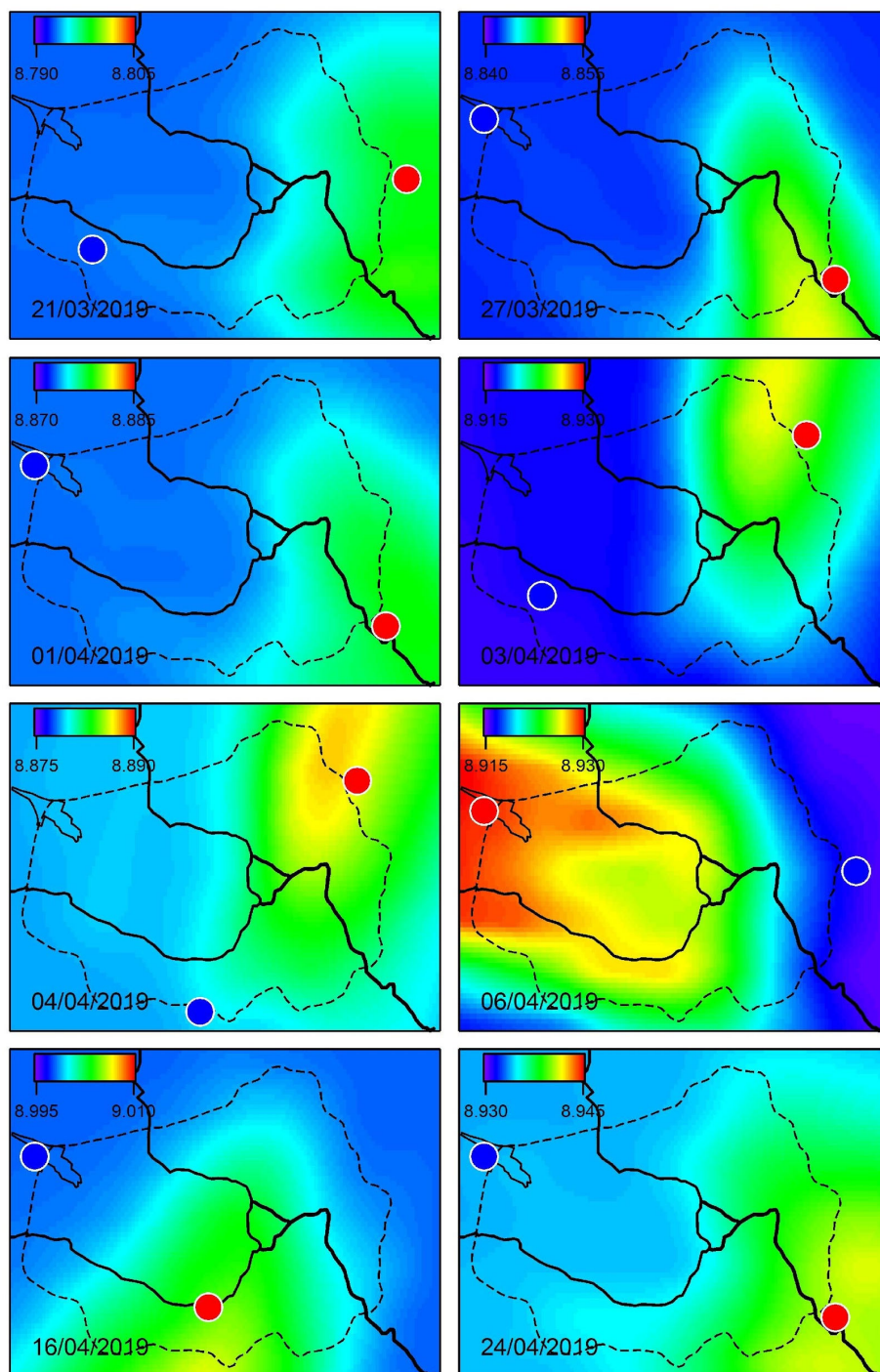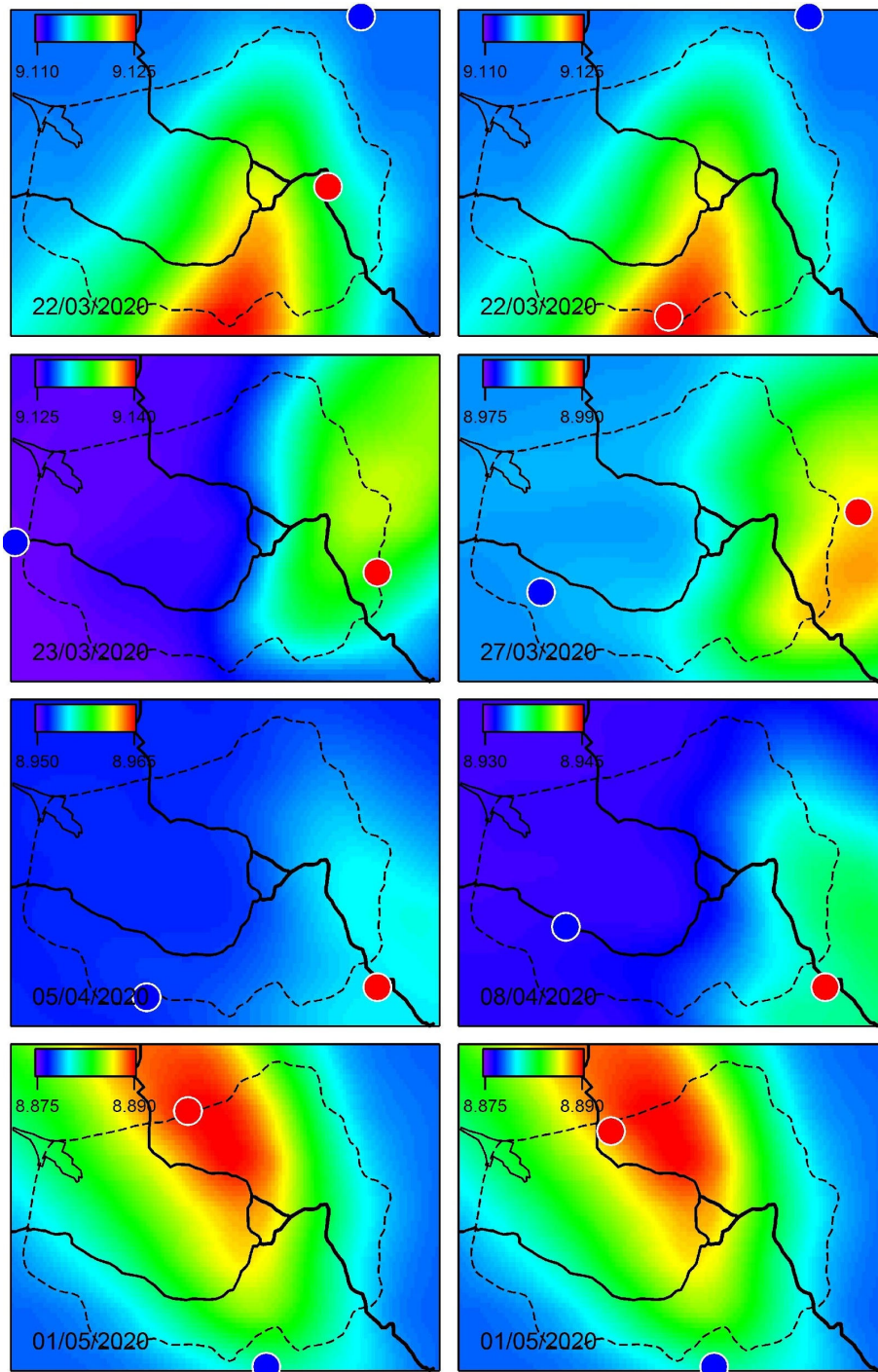
Figure A1: Urban pollution CO_2 plume over St. Petersburg calculated with HYSPLIT model for the days of field campaign in 2019 (10:00 UTC). The colour bar units for TC_{CO2} are 10^{21} cm^{-2}. The blue and red circles indicate the locations of upwind and downwind FTS observations, accordingly.

Figure A2: Urban pollution CO₂ plume over St. Petersburg calculated with HYSPLIT model for the days of field campaign in 2020 (10:00 UTC). The colour bar units for TC_{CO2} are 10^{21} cm⁻². The blue and red circles indicate the locations of upwind and downwind FTS observations, accordingly.

*You should keep in mind that due to the air mass dependencies the diurnal differences do not necessarily reflect changes in the emissions. So knowing the measurement times is critical.

We agree with the referee's comment. Indeed, TCCON and COCCON FTIR observations have high precision and accuracy and usually reveal the CO_2 diurnal cycle with the maximum around the local noon. For our case, it is assumed that the CO_2 diurnal cycle for the background atmospheric conditions could be estimated using FTIR observations at the upwind location where the influence of air pollution is minimal. According to the results obtained during 2019 campaign, the pronounced diurnal variations of CO_2 for upwind locations were observed during only one day (06.04.2019, please see Appendix B, Figure B1). For this day the CO_2 TC had the variation of 0.2%. For other seven days of EMME-2019, the CO_2 diurnal cycle showed significantly lower (or even negligible) amplitudes (see Figure B1). Therefore, relying on the results of mobile campaign of 2019 we assumed that it is quite possible to perform mobile FTIR observations in 2020 using only one EM27/SUN spectrometer (i.e. making FTIR measurements at up- and downwind locations sequentially) even if this approach can introduce certain additional errors.

*2. Which retrieval algorithm was used for analyzing the FTIR spectra?

To analyze the FTIR spectra we used the free software PROFFAST specially developed for COCCON network. We explain this in the new subsection 2.1 "Bruker EM27/SUN FTS and spectra processing" of Section 2 "Methods and instrumentation" that we add to the text:

To process the spectral data we used the free software PROFFAST which had been specially developed for COCCON network (COCCON, 2021; Frey et al., 2019). PROFFAST has been developed by KIT in the framework of several ESA projects for processing the raw data delivered by the EM27/SUN FTS. For the retrievals of total columns (TCs) of target species the following spectral bands are used (Frey et al., 2015; Hase et al., 2016; Frey et al., 2019): 4210-4320 cm^{-1} (target gas – CO, interfering gases – H_2O, HDO, CH_4), 5897-6145 cm^{-1} (target gas – CH_4, interfering gases – H_2O, HDO, CO_2), 6173-6390 cm^{-1} (target gas – CO_2, interfering gases – H_2O, HDO, CH_4), 7765-8005 cm^{-1} (target gas – O_2, interfering gases – H_2O, HF, CO_2), and 8353-8463 cm^{-1} (target gas – H_2O). The retrieval algorithm requires the following input: temperature profile in the atmosphere, pressure at the ground level, and the a priori data on the mole fraction vertical distribution of the atmospheric trace gases. These data are generated by the TCCON network software which ensures their compatibility over the TCCON network (TCCON, 2021). The close agreement of EM27/SUN observations analyzed with PROFFAST with a collocated TCCON spectrometer has been demonstrated in the framework of the ESA project FRM4GHG (Sha, 2020).

*3. Could you show the timeseries of XCO_2 or ΔCO_2 showing FTIR measurements for a typical day or the entire period of measurements? This would help the readers understand the daily variations and enhancements better.

We followed the advise of the referee and now the revised version of the manuscript contains an appendix in which the data series of measured and calculated CO_2 content are shown for the selected campaign days in 2019 and 2020 (Appendix B, Fig. B1 and B2):

Appendix B: The data series of measured and calculated CO_2 content

The upwind and downwind CO_2 total column values acquired from FTIR measurements and HYSPLIT calculations are shown for selected campaign days in 2019 and 2020 in Fig. B1 and B2. The HYSPLIT data are in fact the values of an integrated vertical column in the range of 0-1500 meters (10 altitude layers) calculated with the 15-minute time step. The background level of the CO_2 column is set equal to an average of the FTIR upwind measurements during a day. Note

that in 2020, there were days when the downwind measurements were performed twice, at different locations – on March 22 and May 1 (see Fig. B2).
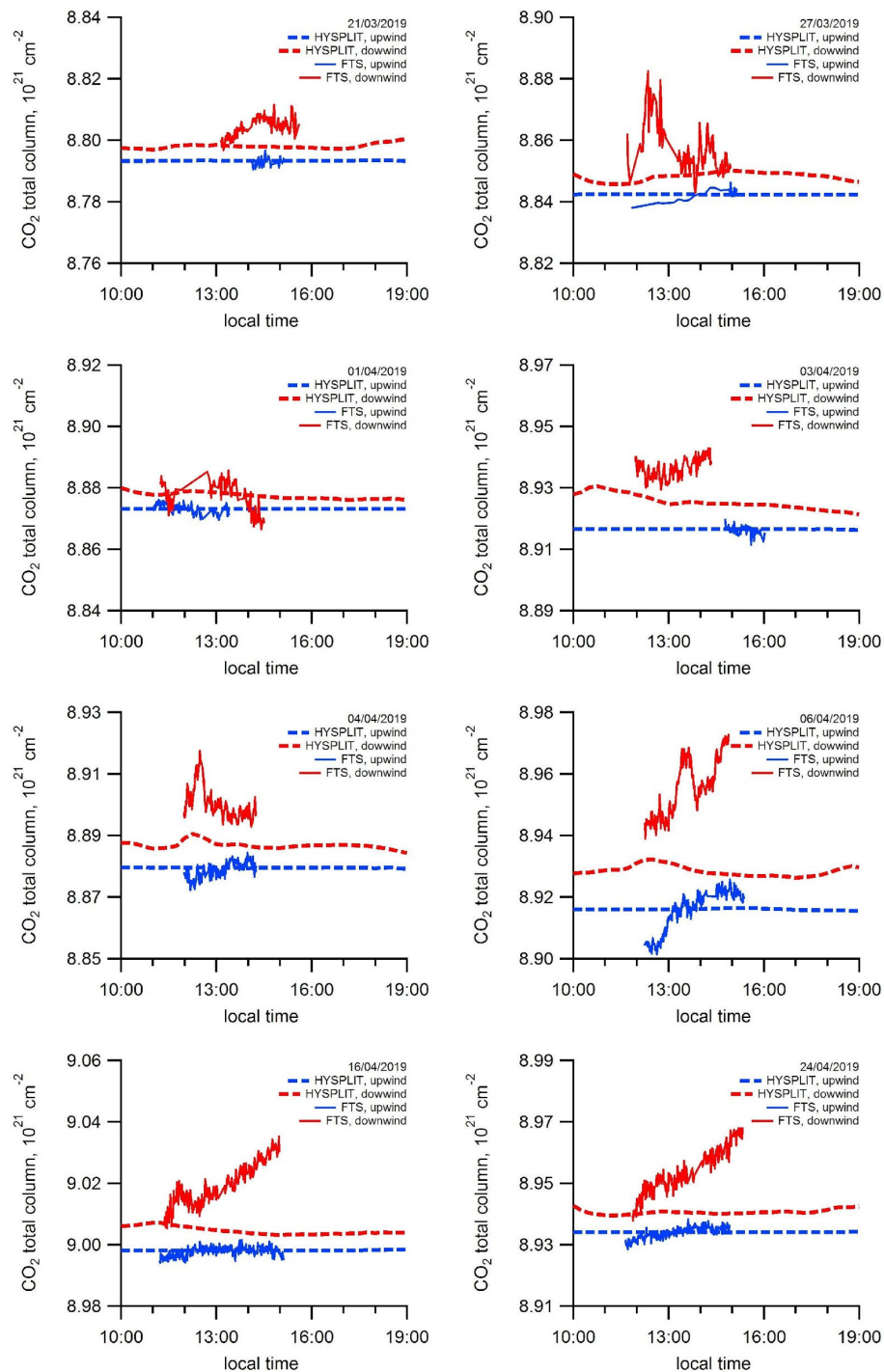

Figure B1: Time series of measured (FTS) and simulated (HYSPLIT, without scaling of the ODIAC emissions data) CO$_2$ total column at the upwind (blue lines) and downwind (red lines) locations for selected campaign days in 2019.
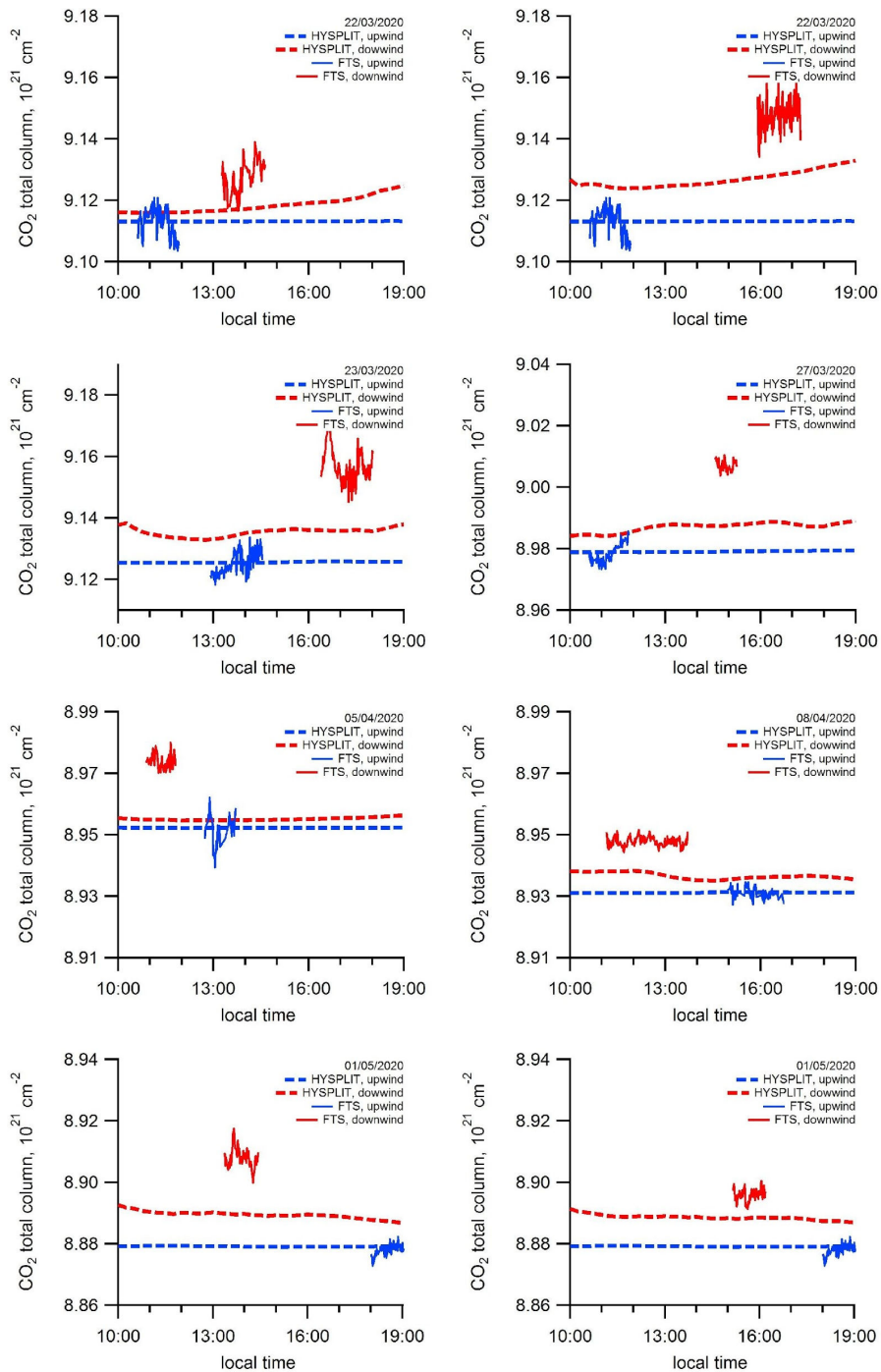
Figure B2: Time series of measured (FTS) and simulated (HYSPLIT, without scaling of the ODIAC emissions data) CO$_2$ total column at the upwind (blue lines) and downwind (red lines) locations for selected campaign days in 2020.

In addition, two examples of HYSPLIT-simulated CO$_2$ plumes and corresponding time series of CO$_2$ total column measurements for the typical days of experiments in 2019 and 2020 are presented in Figs. 5 and 9:
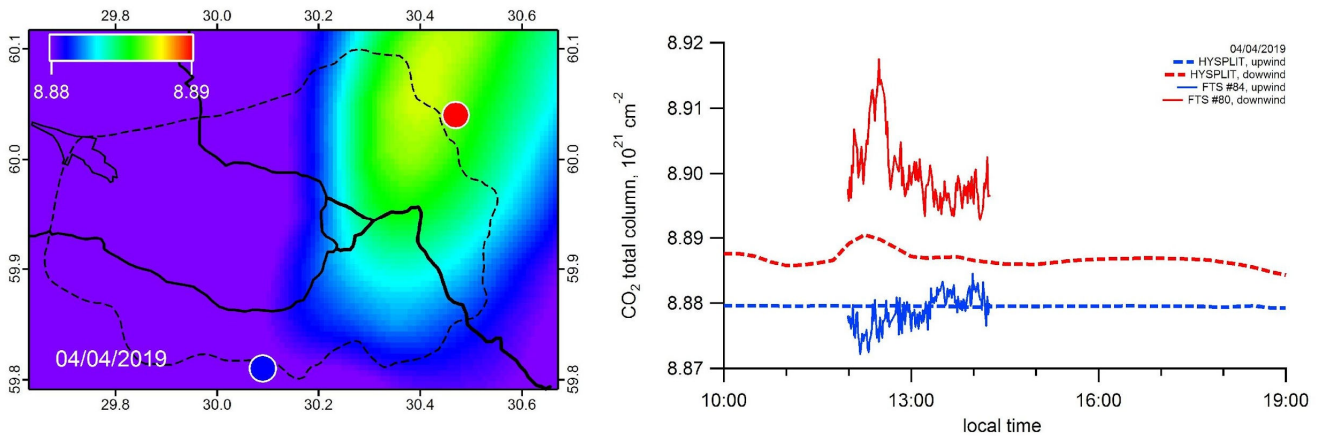
Figure 5: Left panel: Urban pollution CO_2 plume over St. Petersburg calculated by HYSPLIT model for April 4, 2019 (10:00 UTC). The colour bar designates the CO_2 total column in units 10^{21} cm^{-2}. The blue and red circles indicate the locations of upwind and downwind FTS observations, accordingly. Right panel: Time series of measured (FTS) and simulated (HYSPLIT, without scaling of the ODIAC emissions data) CO_2 total column at the upwind (blue lines) and downwind (red lines) locations for the same day.
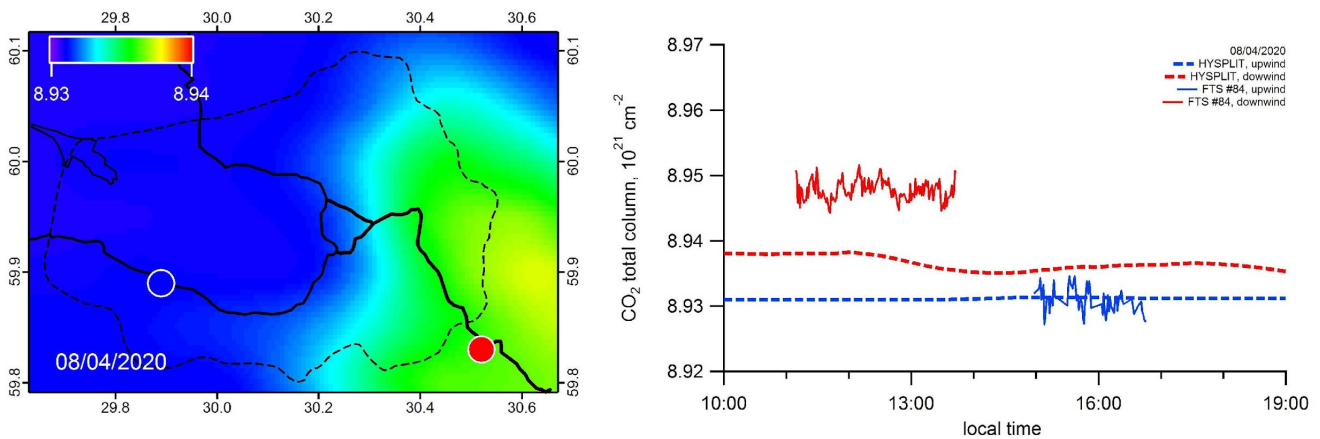

Figure 9: Left panel: Urban pollution CO_2 plume over St. Petersburg calculated by HYSPLIT model for April 8, 2020 (10:00 UTC). The colour bar designates the CO_2 total column in units 10^{21} cm^{-2}. The blue and red circles indicate the locations of upwind and downwind FTS observations, accordingly. Right panel: Time series of measured (FTS) and simulated (HYSPLIT, without scaling of the ODIAC emissions data) CO_2 total column at the upwind (blue lines) and downwind (red lines) locations for the same day.

*4. How the two FTIR instruments compare to each other when measuring side by side? How are you accounting for potential instrument biases?

Yes, we certainly took care of this issue. To give some details we have added the following text to subsection 2.1 "2.1 Bruker EM27/SUN FTS and spectra processing":

In order to obtain a reliable value of the CO_2 emission for St. Petersburg, it is necessary to eliminate possible systematic error caused by the instrument bias. This goal was reached by carrying out a cross-calibration of the instruments. In April-May 2019 both instruments passed a four day cross-calibration. The comparison of side-by-side measurements of X_{CO2} by FTS#80 and FTS#84 allowed determining calibration factor which was used for converting X_{CO2} measured by FTS#80 to the scale of FTS#84. Detailed information about side-by-side calibration of FTIR-spectrometers is given in the paper by Makarova et al. (2021).

*5. You mentioned you have excluded a pixel from ODIAC priors because the emissions seemed to be an outlier. What if it's the emissions from a power plant(s) that was misattributed to a wrong location? It is expected that industrial sources and power plants stand out as they are point sources with significant amount of emissions. If you any valid reason to modify emissions from that pixel you should clearly state that you are not using ODIAC but a modified version of ODIAC.

Indeed, power plants and industrial enterprises normally manifest themselves as point sources of strong emission. However, we cannot confidently attribute this particular ODIAC pixel to any source of this type, since there is no such object near it. There are about a dozen of large thermal power plants on the territory of St. Petersburg, but all of them appear to be rather far from this location. Given the reviewer's doubts, we have studied this issue more thoroughly. Despite the lack of published data on anthropogenic CO_2 emissions at the city scale, we were able to explore detailed reports from municipal inventories of stationary air pollution sources (unpublished, but available on request). According to the inventories of NO_x, CO, SO_2, NH_3, VOC and PM10 pollutants, there are no stationary objects of an extreme emission close to the point of our interest. Thus, we feel confident to smooth out this outlier when we configure emissions to run the HYSPLIT model. However, we do not consider it necessary to mark this as an "ODIAC modification". Removing one pixel from several thousand in the database does not mean that it is modified significantly. In any case, it is not possible to implement ODIAC directly into HYSPLIT, without adaptation. Nevertheless, we have added a few words of justification to the appropriate place in the text:

Normally, power plants and industrial enterprises manifest themselves as point sources of strong emission. However, we cannot confidently attribute this particular ODIAC pixel to any source of this type, since there is no such object near it. There are about a dozen of large thermal power plants on the territory of St. Petersburg, but all of them appear to be rather far from this location. Despite the lack of published data on anthropogenic CO_2 emissions at the city scale, we were able to explore detailed reports from municipal inventories of stationary air pollution sources (unpublished, but available on request). According to the inventories of NO_x, CO, SO_2, NH_3, VOC and PM10 pollutants, there are no stationary objects of an extreme emission close to the point of our interest. Thus we feel confident to smooth out this outlier and replace it by the value averaged over the neighboring ODIAC pixels.

*6. For comparisons of in-situ measurements and HYSPLIT model results you are using a fixed background of 415 ppm. Given the day to day variations in the CO_2 levels it's not very reasonable to use a fixed value for the entire period of the analysis unless you bring proof that this was the case for St Petersburg during the campaign.

As it was mentioned in the article (subsection 3.3 in the original manuscript version, *"Simulations of ground-level CO_2 concentrations"*), routine measurements of the CO_2 surface concentrations have been carried out since 2013 using a gas analyzer Los Gatos Research GGA 24r-EP located at the

atmospheric monitoring station of Saint-Petersburg State University in Peterhof (59.88°N, 29.83°E). 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. Thus, for testing HYSPLIT model setup for the St. Petersburg region, we used background value of the CO_2 surface concentration near Peterhof during the 2019 EMME measurement campaign – from March 20 to April 30, 2019 (Makarova et al., 2021). We used the background value of the surface concentration equal to 415 ± 2 ppmv, which is the mean value of diurnal minima during the campaign. We have added this explanation to the revised manuscript (subsection 2.4 "Test simulations of ground-level CO_2 concentrations"):

The background value of the surface concentration is taken as 415 ppmv based on the local measurements (415 ± 2 ppmv is the mean value of diurnal minima during the campaign from March 20 to April 30, 2019).

*7. It will be useful if you also plot the one on one curve for HYSPLIT vs in-situ measurements from which you found out a scaling factor.

We have changed Figure 4 to show the regression plot of HYSPLIT vs in-situ CO_2 measurements, as suggested by the referee.
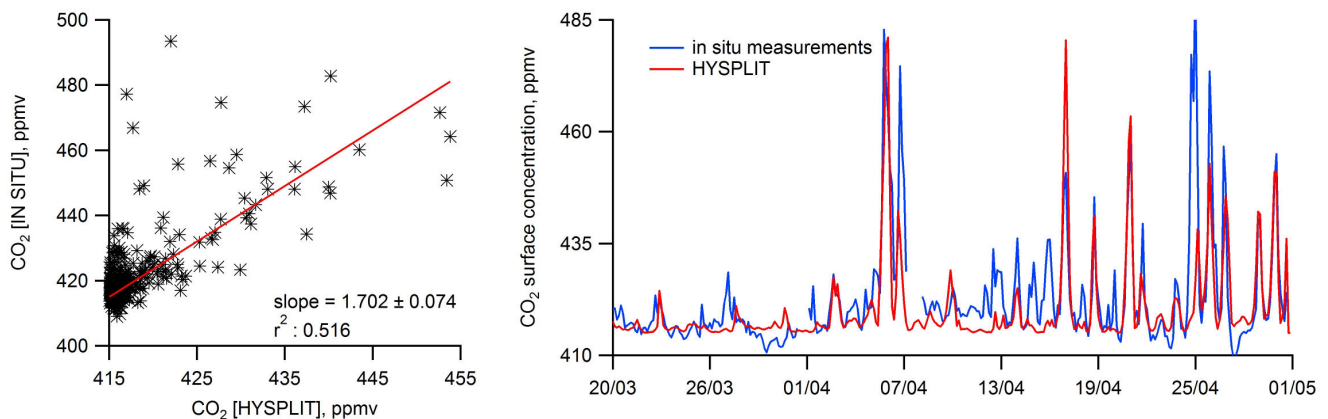

Figure 4: Comparison of the HYSPLIT simulations and the in situ measurements of surface CO_2 concentration in Peterhof (59.88° N, 29.82° E) in March-April 2019. Left panel: The values of surface CO_2 compared with the results of HYSPLIT simulations without scaling of the ODIAC emissions data. Right panel: 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.

*8. Please describe in details how did you estimate fluxes using the mass balance method. Bring equations if necessary.

In fact, calculations, which use the mass balance approach, are only supplementary in the present paper. It was the subject of our earlier study published by Makarova et al. (2021) with all the necessary equations presented there. The details of the flux estimates obtained by mass balance approach are definitely out of the scope of the current study. However, we have added the following brief explanations to the revised text of our manuscript:

Basically, the mass balance approach was adopted in the form of a one-box model, and the area flux F was calculated using the following equation:

$$F = \frac{\Delta CO_2 \cdot V}{L}$$

(2)

where F is the CO_2 area flux, ΔCO_2 is the difference between the downwind and upwind FTIR measurements, V is the mean wind speed and L is the mean path length of an air parcel which goes through the urban area (Chen et al., 2016).

*9. How are you computing column ΔCO2 using HYSPLIT? Do you take an integral over the vertical layers of 1m-1500m?

Exactly so. We take an integral over the CO_2 concentrations calculated using HYSPLIT for 10 vertical layers in the range of 0-1500 meters. This is what we write in the original text of our manuscript (beginning of Section 4 in the original manuscript version, "*Evaluation of integrated CO_2 emissions from field FTIR measurements*"): "*HYSPLIT model was configured to simulate CO_2 concentrations at 10 altitude levels (0-1500 m), which were then integrated to obtain the CO_2 column in the boundary layer. The differences between the results of FTIR measurements of the CO_2 TC inside and outside the pollution plume (ΔCO_2) were compared with the differences in the CO_2 column in the boundary layer simulated by HYSPLIT at the corresponding locations*".

*Detailed comments:

*Line 26: From the value in brackets I imply that emission estimates during the lockdown period was higher than the rest but the last sentence of abstract is suggesting there was a 8% decrease. Please clarify!

Indeed, this part of the text is not entirely clear. In fact, the strict allocation of the period of influence of the quarantine based on the formal terms of its introduction is not quite correct, since some decline in business activity could have started even before the official introduction of restrictive measures. Therefore, in the revised version of our manuscript, we abandoned this approach and now compare the results of all measurements in 2020 with the results of all measurements in 2019. We have made the appropriate changes to the Annotation and to Section 4 "Summary and conclusions":

The most important finding of the present study based on the analysis of two observational campaigns is a significantly higher CO_2 emission from the megacity of St. Petersburg as compared to the data of municipal inventory: ~75800±5400 kt yr^{-1} for 2019, ~68400±7100 kt yr^{-1} for 2020 versus ~30000 kt yr^{-1} reported by official inventory. The comparison of the CO_2 emissions obtained during the COVID-19 lockdown period in 2020 to the results obtained during the same period of 2019 demonstrated the decrease in emission of 10% or 7400 kt yr^{-1}.

*Line 42: Please consider adding a proper citation.

The following reference has been added: UN: United Nations, Department of Economic and Social Affairs, Population Division: World Urbanization Prospects 2018: Highlights. https://population.un.org/wup/Publications/Files/WUP2018-Highlights.pdf, last access 29 March 2021.

*Line 43: Do you mean anthropogenic CO_2 emissions? Also having total fossil fuel consumption is not enough to estimate anthropogenic emissions. There are emissions

In response to this comment of the reviewer, we have rewritten the mentioned sentence:

The vast majority of anthropogenic CO_2 emissions in developed countries are associated with the burning of fossil fuels (FF) and can be estimated with good accuracy on the basis of the total fuel consumption.

*Line 77 and 78: Please bring proper citations for both official and unofficial population estimates.

The following reference containing the official information on the population of St. Petersburg has been added:

St. Petersburg Center for Information and Analytics: Report on Demographic monitoring in St. Petersburg: Q3 2020, https://www.gov.spb.ru/helper/new_stat/, 2020, (in Russian).

The reference containing the unofficial estimates of the population of St. Petersburg has been added:

Shevlyagina M. The real population of St. Petersburg exceeds 7 million people, https://spbdnevnik.ru/news/2020-02-27/realnoe-naselenie-peterburga-prevyshaet-7-millionov-chelovek, last access 29 March 2021 (in Russian).

*Line 88: How much is the contribution from transport in St Petersburg? It's important since you are mentioning transport emission changes during lock down period later in the text.

As we noted later in the text of the manuscript (subsection 4.2 in the original manuscript version, *"The results of EMME-2020 and comparison with EMME-2019"*), official data on the contribution of transport to the total CO_2 emissions in St. Petersburg are not available:

Since we have no official data on the CO_2 emissions by traffic at our disposal, we used the average estimate for European countries, according to which the contribution of traffic to total emission constitutes 30% (European Parliament News, 2020).

According to the report prepared by the analytical center of Gazprom public joint stock company (Gazprom, 2021) the total CO_2 emission from motor vehicles in St. Petersburg was of ~10 Mt/year in 2014. This value is about one third of the total CO_2 emission of St.Petersburg (~30 Mt/year) reported by the municipal Environmental Committee (Serebritsky, 2018). This supports our assumption of a 30% contribution of transport to total CO_2 emissions.

Gazprom: The ecological effect of the conversion of motor vehicles to gas-powered fuel in the regions of the Russian Federation, https://www.gazprom.ru/f/posts/22/538143/atlas-ecology-effect-gas-transport.pdf, last access 30 March 2021 (in Russian).

Serebritsky, I.A., (Ed.): The Report on Environmental Conditions in St. Petersburg for 2017, https://www.gov.spb.ru/static/writable/ckeditor/uploads/2018/06/29/Doklad_EKOLOGIA2018.pdf, 2018 (in Russian).

*Line 113: As mentioned earlier please briefly describe the campaign in a separate paragraph don't assume audience have read the other paper. Then state what are the

Following the reviewer's suggestion, the new Section 2 of the revised manuscript ("Methods and instrumentation") contains the subsection 2.1 "Bruker EM27/SUN FTS and spectra processing" which describes the instrument and spectra retrieval in detail:

Bruker EM27/SUN (Gisi et al., 2012; Frey et al., 2015, Hase et al., 2016) is a portable robust FTS having low spectral resolution of 0.5 cm^{-1}. It was designed for accurate and precise ground-based observations of CO_2, CH_4 and CO column-averaged abundances (X_{CO2}, X_{CH4} and X_{CO}) in the atmosphere. These FTIR spectrometers were used to build the COCCON network (COCCON, 2021; Frey et al., 2019). EM27/SUN is equipped with a Camtracker, a solar tracking system developed by KIT (Gisi et al., 2011). A Camtracker consists of an altazimuthal solar tracker, a USB digital camera and "CamTrack" software which processes an image acquired by a camera and controls the tracker's movement. EM27/SUN FTS is designed on the basis of a robust RockSolid™ interferometer having high thermal and vibrational stability; the detailed description of the instrument is given by Gisi et al. (2012). Therefore, this type of instruments is being successfully implemented for setting up fully automated stationary city network MUCCnet (Munich Urban Carbon Column network, Dietrich et al., 2021) and for performing a number of mobile campaigns (Klappenbach et al., 2015; Luther et al., 2019; Makarova et al., 2021).

In our study, we used the dual-channel EM27/SUN with quartz beamsplitter. Additionally, two detectors allow observing X_{CO} and future improvements of the X_{CO2} retrieval (Hase et al., 2016). FTS registers an interferogram which is the Fourier transform of the infrared spectrum of direct solar radiation. The processing of data acquired by EM27/SUN spectrometer consists of the following stages:
- deriving spectra from raw interferograms including a DC-correction and quality assurance procedures (Keppel-Aleks et al., 2007);
- deriving O_2, CO_2, CO, H_2O, and CH_4 total columns (TCs) from FTIR spectra by scaling a priori profiles of retrieved gases (Frey et al., 2019; COCCON, 2021).

To process the spectral data we used the free software PROFFAST which had been specially developed for COCCON network (COCCON, 2021; Frey et al., 2019). PROFFAST has been developed by KIT in the framework of several ESA projects for processing the raw data delivered by the EM27/SUN FTS. For the retrievals of total columns (TCs) of target species the following spectral bands are used (Frey et al., 2015; Hase et al., 2016; Frey et al., 2019): 4210-4320 cm^{-1} (target gas – CO, interfering gases – H_2O, HDO, CH_4), 5897-6145 cm^{-1} (target gas – CH_4, interfering gases – H_2O, HDO, CO_2), 6173-6390 cm^{-1} (target gas – CO_2, interfering gases – H_2O, HDO, CH_4), 7765-8005 cm^{-1} (target gas – O_2, interfering gases – H_2O, HF, CO_2), and 8353-8463 cm^{-1} (target gas – H_2O). The retrieval algorithm requires the following input: temperature profile in the atmosphere, pressure at the ground level, and the a priori data on the mole fraction vertical distribution of the atmospheric trace gases. These data are generated by the TCCON network software which ensures their compatibility over the TCCON network (TCCON, 2021). The close agreement of EM27/SUN observations analyzed with PROFFAST with a collocated TCCON spectrometer has been demonstrated in the framework of the ESA project FRM4GHG (Sha, 2020).

To our opinion, this comment made by the referee repeats in a certain way several previous comments relevant to description of the measurement campaigns and data processing algorithms. In fact, the answer to this comment of the referee has already been given above when we introduced appendixes and the subsections of the new Section 2 "Methods and instrumentation". In addition, in accordance with the referee's suggestion, we have moved the mentioned part of the text to Section 4 "Summary and conclusions".

For the sake of certainty, we have rephrased the text as follows:

It should be noted that the value of ΔXCO_2 depends not only on the integral emission of the source, but also on its type (point, linear or area), the geometry of the field experiment (location of observations relative to the pollution plume) and on the meteorological situation during the measurements.

For clarity, we have rewritten this part of the text:

The nominal latitude/longitude size of the ODIAC data pixel is 30 arcseconds (Oda and Maksyutov, 2011), which defines a global spatial resolution of about 1 km × 1 km. Since the length of a degree of longitude changes with the latitude, the pixel size for St. Petersburg (~60° N) is smaller and equals to 0.93 km × 0.46 km (0.43 km^2).

No, this value (32529 kt) is an estimate of CO_2 annual emission coming from the original ODIAC database (see Section 2.2 "A priori data on FF CO_2 emissions"). If we smooth out the ejected pixel, as explained in the paper, we get 29524 kt. Thus, the difference is less than 10%, and this can hardly explain the observed discrepancy between the model and the measurements.

We now have this information in the new tables, 1 and 2 (see above). So, for clarity, we add the reference to one of these tables here:

For the sake of comparison, the simulation results and measurement data were averaged over time periods of field observations (the duration of each experiment is given in Table 1).

In the revised version of the manuscript full details of both field campaigns are given in Tables 1 and 2 for 2019 and 2020, respectively. In addition, we provide an appendix showing the data series of measured and calculated CO_2 content for the selected campaign days in 2019 and 2020 – Appendix B, Fig. B1 and B2 (see above). Thus, we refer the reader to these plots in the appendix:

> The error bars indicated in Fig. 5a as boxes are in fact the variations of ΔCO_2 obtained as standard deviation of observations and simulations within one observational series (see Appendix B, Fig. B1).

*Line 251. The sentence is unclear. Please consider rewording.

The sentence was reworded as follows:

> An earlier analysis of the results of the EMME-2019 measurement campaign focused in particular on inferring the area fluxes of urban CO_2 emissions from St. Petersburg. In order to achieve this goal, the simplified mass balance approach was applied to the observed CO_2 enhancements (ΔCO_2) which were attributed to the accumulation of pollution during the air mass movement on its way from the upwind side to the downwind side of the megacity (see Makarova et al., 2021 for full details).

*Line 294: The sentence is unclear. Please consider rewording or adding more explanation.

The sentence was reworded as follows:

> Our confident expectation to detect the transport contribution is based on the high sensitivity of FTIR measurements of XCO_2 using EM27/SUN spectrometers and COCCON methodology.

*Line 302: Please specify in detail which days were excluded from the analysis and how many days are used after the exclusion. As mentioned earlier a table would be helpful.

We add the following explanation to the text here:

> In general, for these reasons (including unfavorable weather conditions and wrong location of FTIR measurement points), data from only a few experiments were excluded: No.8 on April 18, 2019, No.10 on April 25, 2019, No.11 on April 30, 2019 (see Table 1) and No.4 on March 27, 2020 (see Table 2).

We believe that having Tables 1 and 2, as well as the figures in Appendices A and B, a reader has now complete information about the days and time, and the location of all our observations.

*Line 359: Please explain in more details why that's the case. From what I understand the in-situ measurement site is in a fairly remote location but the FTIRs have been deployed in the city centre and closer to large sources. So there are other variables that might play a role here!

Let us explain here. Not only the in situ measurements in Peterhof, but also all mobile observations were planned in such a way that the FTIR measurement points were located at a distance from any local sources of heavy pollution. The main idea was to detect the integral CO_2 enhancement as the

difference between downwind and upwind observations. In this sense the Peterhof site may also appear inside the urban plume and get the high levels of CO_2 surface concentration. However, these surface CO_2 enhancements obviously lack the contributions of elevated emission sources being well mixed in the upper altitude layers. This is not the case for FTIR measurements which probe the column amount of CO_2 and not only the surface layer. We believe that the revised version of our manuscript with new plots and maps makes this clear and needs no further explanation.

*Figure 1 and 9: Please add latitude and longitude coordinates to the map.

Coordinates have been added to the map.

*Figure 3: Please add latitude and longitude coordinates to the bottom map.

Coordinates have been added to the bottom map.

*Figure 6: Please add latitude and longitude coordinates to the map. Also please specify the location of upwind instrument for 2019. It would also be helpful to specify the dates at each location.

Coordinates have been added to the maps in Figure 6 (Figure 7 in the revised version of the manuscript). In order not to overload this figure with information, details about the location of the instruments for each day are presented in separate figures provided in the Appendix A.

*Figure 7: Are the ODIAC area flux values shown the average over the entire domain or the average along the HYSPLIT back-trajectory?

The ODIAC area flux values are the average along the trajectories shown on the map in Figure 6 (Figure 7 in the revised version of the manuscript). We explain this in the text:

> *Schematically, the air trajectories corresponding to the 2019 FTIR measurement locations are shown in Fig. 6. These trajectories were simulated as backward trajectories by the HYSPLIT model in the boundary layer of the atmosphere. The resulting values of anthropogenic CO_2 area fluxes calculated by integrating the ODIAC data along the trajectories presented in Fig. 6, are shown in Fig. 7 in comparison with the experimental estimates by Makarova et al., 2021. As in the study by Makarova et al., 2021, the width of the air paths was assumed to be 10 km*

We also specify this in the caption to Figure 7 (Figure 8 in the revised version of the manuscript):

> *Figure 7: The CO_2 area flux (F_{CO2}) obtained on the basis of the mass balance approach (EMME-2019) compared to the CO_2 area flux derived from scaled ODIAC data. <u>The calculations are made for the trajectories shown in Fig. 6.</u> Dots are connected by lines for illustrative purposes only.*