

The reply to the anonymous referee #2 (RC2)

We are thankful to the referee for the very detailed analysis of our study. We agree with almost all comments and 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**.

The paper is well written, with good language and nice, instructive graphs in most cases.

We are grateful to the referee for the positive assessment of our manuscript.

It is claimed that the objective of the paper is to provide emission numbers for Sankt Petersburg. However a significant, and in my mind, to big part of the paper describes the general methodology with complementary data. The abstract is rather long and detailed, and it should be made more concise with focus on the results. The main body is too detailed for a scientific paper: a) The Modis data is not relevant since it is not actively used, b) Remove nice photos of StPetersburg, c) In the introduction, there is a lot of explanation about different variants of obtaining windspeed and effective path, but this is not used in any significant extent in the results; this should be shortened.

We agree with the referee's statements. However, to our opinion, the details of the experiment can be helpful for better understanding and analysis of the obtained results. Therefore we decided not to remove the experiment details completely or to shrink the corresponding part of the manuscript, but to move these details to the Appendix. We made the following changes in the paper:

- 1) Figure 4 containing MODIS images has been moved to Appendix A;
- 2) Figure 3 has been removed from the revised version of the manuscript;
- 3) Part of the information on the EMME-2019 observation details (including Table 1), the overview of meteorological data for the days of the field campaign (including Table 2), and the analysis of wind speed and the wind direction for the days of the field campaign based on the different data sources (including Table 3) were also moved to Appendix A.

If I understand right, the methodology is the same as used in other campaigns (Berlin). In the introduction or elsewhere an overview about the other studies should be added with discussion on how comparable this study is to the other ones in terms of methodology and results . E.g. was effective path used by other studies.

Yes, the esteemed referee is right. In the introduction section of the original manuscript it was indicated: "The idea and the methodology of EMME experiment was based mainly on the studies by Hase et al. (2015), Ionov and Poberovskii (2015), Chen et al. (2016) and Viatte et al. (2017)". Following the advice of the referee we added the following text:

... 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. Vogel et al. (2019) investigated the Paris megacity emissions of CO₂ by coupling the COCCON observations and atmospheric transport model framework (CHIMERE-CAMS) simulations.

... De Foy et al. (2007), Mellqvist et al. (2010), Johansson et al. (2014), and Kille et al. (2017) have applied mobile FTIR (Solar Occultation Flux technique) and mobile DOAS techniques to the large scale flux measurements.

In Eq 1 you calculate the flux using total column (needed to get the right unit).

We have made the necessary changes in section 4.2 Mass balance approach for area flux estimation. The new version of this section which includes explicit indication of the units is given below:

The estimation of the area fluxes F was obtained on the basis of a mass balance approach implemented in the form of a one-box model. Box models are a widely used technique for the evaluation of urban and other emission fluxes (Hanna et al., 1982; Reid and Steyn, 1997; Arya, 1999; Zinchenko et al., 2002; Zimnoch et al., 2010; Strong et al., 2011; Hiller et al., 2014a; Chen et al., 2016; Makarova et al., 2018). In our case the following equation for the calculation of area flux was used:

$$F_j(t_k) = \frac{\Delta_{TC}(t_i) \cdot V_j(t_i)}{L_j(t_i)} \cdot k, \quad (2)$$

where F (unit: $t \text{ km}^{-2} \text{ yr}^{-1}$) is the area flux, t_i denotes the day of a single field experiment in the frame of the observational campaign. It should be emphasized that we used the steady-state approximation for all involved processes within the duration of a single field experiment, so Δ_{TC} (unit: molec. m^{-2}) is the mean TC difference between downwind (TC_d) and upwind (TC_u) observations $\Delta_{TC} = TC_d - TC_u$, V (unit: m sec^{-1}) is the mean wind speed, and L (unit: m) is the mean length of a path of an air parcel which goes through the urban territory of St. Petersburg agglomeration. The k coefficient converts the value of area flux from (unit: $\text{molec. m}^{-2} \text{ sec}^{-1}$) to (unit: $t \text{ km}^{-2} \text{ yr}^{-1}$):

$$k = \frac{m_{gas} \cdot 31536 \cdot 10^6}{N_A}, \quad (3)$$

where m_{gas} is the molecular mass of the target gas (unit: kg mol^{-1}), N_A – Avogadro constant (unit: mol^{-1}), $31536 \cdot 10^6$ - the coefficient that converts the value of area flux from (unit: $\text{kg m}^{-2} \text{ sec}^{-1}$) to (unit: $t \text{ km}^{-2} \text{ yr}^{-1}$). The data for the wind speed and the wind direction were taken from different sources of meteorological information (see section 4.3), and these sources are identified as j in Eq. 2. So, as a result, we obtained the set of values of $F(t)$ for each of the meteorological data sources and for each day of field measurements. We note that below we will use the units $t \text{ km}^{-2} \text{ yr}^{-1}$ for the values of $F(t)$.

You also introduce Xgas (I assume against total pressure). When do you use Xgas in the calculation? Is it only to show thing quantitatively? I assume in most cases te pressure is the same for up and downwind site ? Add in the text a definition of Xgas (not know for everyone) and describe what is your purpose here for showing it?

Please, see the answer to this comment below (the answer to referee's comment to P8, row 128).

For the wind used in the final results the authors rely on the Hysplit model, which in turn is based on a global model (NCEP) for the wind. The authors argue that the use of data from this model provides less variability in the final results. I argue that the wind variability is less

for the Hysplit data than for real measurements, since it is large domain model, and Hysplit will therefore artificially smooth the wind data. This should be better discussed by the authors.

We agree with the referee's statement that "wind variability is less for the Hysplit data than for real measurements ... and Hysplit will therefore artificially smooth the wind data". Nevertheless, to our opinion, HYSPLIT cannot be classified as a "...large domain model...". Following the advice of the referee, we presented our arguments in the extended discussion in the new version in Section 4.4:

We selected HYSPLIT as one of the sources of the wind data since HYSPLIT is a widely used modelling system for the simulation of air parcel trajectories and the dispersion processes in the atmosphere which was tested in a lot of studies (HYSPLIT publications can be found using the following links: <https://www.arl.noaa.gov/hysplit/hysplit-publications-meteorological-data-information/>). Stein et al. (2007) noted that *Grid models are the best-suited tools to handle the regional features of these chemicals. However, these models are not designed to resolve pollutant concentrations on local scales. Moreover, for many species of interest, having reaction time scales that are longer than the travel time across an urban area, chemical reactions can be ignored in describing local dispersion from strong individual sources making Lagrangian and plume-dispersion models practical.* Stein et al. (2007) classify HYSPLIT as a local model which provides *the more spatially resolved concentrations due to local emission sources.* Therefore, for modelling of the evolution of the St.Petersburg plume we used the HYSPLIT model as a tool which perfectly fits the scale of considered atmospheric processes. This was also the reason for using HYSPLIT as the source of the wind data.

The authors present their flux estimation based on modelled effective path. Such an exercise provides useful data but it is hard for the reader to understand how the data was produced and its errors, since the data represents a combination of measurements and model. I suggest presenting also the purely measured data based on a constant path. For the effective path the authors claim they made a land use analysis and they refer to a public web site but there little information given in the paper and it is hard for the reader to understand the assumptions made here. For instance, I am missing an explanation about what are the hypothesis about the detailed emission source categories and differentiation between species (CO₂, CH₄, NO₂). The species above originate from different emission source categories; e.g CH₄ could partly come from the waterways (sewers and water canals) and pipelines rather than mobile and fixed combustion sources which are relevant for CO₂ and NO₂. This will make the effective path species dependent. The emissions from water ways could also be impacted by windspeed. I suggest adding a graph for the landuse model and include the model as complementary material for this paper.

Addressing this issue, in the revised version of the paper we present the values of area flux calculated using constant path length and the description of the land use model. The results obtained with a constant path length are given in Table B1 (please see below) in the Appendix B.

Table B1. 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 using constant path length approach.

Area flux	EMME		In situ measurements
	(9 days)	(4 days)	
1	2	3	4

CO ₂ , kt km ⁻² yr ⁻¹	96 ± 25	99 ± 17	32 ± 27
CH ₄ , t km ⁻² yr ⁻¹	151 ± 82	213 ± 57	95 ± 64
CO, t km ⁻² yr ⁻¹	276 ± 117	385 ± 97	71 ± 40
NO _x , t km ⁻² yr ⁻¹	74 ± 30	-	-

The land use model that was developed for the computation of the variable path length is presented in Fig.6 (former Fig.8):

In Fig. 6 these land use classes are shown in different colours: blue for the water bodies, grey for the residential buildings/industrial areas, green for the parks and forests. Effective path length is calculated as a sum of elementary paths through the urbanized grid pixels which contain residential buildings, industrial areas, and roads/highways. Pixels containing water bodies, swamps, and parks are excluded from the variable path calculations. Similar approach was implemented by Hase et al. (2015). The total urbanized area of the St.Petersburg agglomeration according to the developed land use classification occupies the area of 984 km² while the official area of the entire St.Petersburg is of 1439 km². The target gases can originate from different emission source categories, i.e. CH₄ could partly come from the waterways (sewers and water canals), wetlands and pipelines rather than mobile and point combustion sources which are relevant to CO, CO₂ and NO₂. The EMME-2019 was carried out during March-April when water bodies and earth surface were fully or partly covered by ice and snow (see Appendix A, Fig. A1), and soils were still frozen. Therefore we suggest that the CH₄ emission from the excluded pixels (water bodies, swamps, parks, and forests) was negligible in comparison to other anthropogenic sources (landfills, pipelines, etc.) which are distributed over the urbanized pixels.

We generally agree with the statement that “the emissions from water ways could also be impacted by windspeed” but this effect is not expected to be critical since water bodies were covered by ice and snow.

As it was mentioned above, for the revised version of the manuscript we computed the urbanized area of St.Petersburg agglomeration according to the land-use classification that was developed in order to estimate the effective path lengths. The total urbanized area of the agglomeration occupies 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, corresponding 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 (Serebritsky, 2018, 2019)	63-252 (London, Lee, 2015) 13- 300 (Norfolk, Marr, 2013)

The NO₂ DOAS data are explained very briefly wrt to methodology and results. Did you use the same methodology as for the other species, even though you measure in a full circle around town. I suggesting describing the methodology in a better way and results. Did you use the NO₂ data to correct the FTIR measured data, if so clarify.

A detailed description of our DOAS measurements can be found in the references provided in the manuscript (Ionov and Poberovskii 2012, Ionov and Poberovskii 2015, Ionov and Poberovskii 2017, Ionov and Poberovskii 2019). We would not like to increase the size of the manuscript by describing the methodology in every detail. However, as a response to the referee's comment, in the revised version we added the following text to Section 4:

Basically, the DOAS algorithm derives the NO₂ atmospheric content by fitting a reference NO₂ absorption cross-section to the measured zenith scattered radiance. The effective or slant column density (SCD) of NO₂ is retrieved in the 425-485 nm fitting window. SCD is converted then to vertical column density (VCD) by means of so-called air mass factor AMF (VCD=SCD/AMF), pre-calculated with a radiative transfer model (RTM). The spatiotemporal variations of stratospheric NO₂ are negligible compared to these in a polluted

troposphere. Consequently, the variations of NO₂ vertical column observed in the data of our mobile DOAS measurements are related to NO₂ pollution in the boundary layer (below ~1.5 km).

The primary purpose of mobile DOAS NO₂ measurements was a real-time verification of the pollution plume location with respect to the original HYSPLIT dispersion forecast. By means of this approach, the actual evolution of plume was monitored to adjust the FTIR field measurement positions, if necessary. We do mention this in the manuscript: "The real-time corrections of the FTIR operation sites were performed depending on the *actual evolution of the megacity NO_x plume as detected by the mobile DOAS observations*" (lines 35-36 of the Abstract, orig. version), and "The concept of EMME is based on remote measurements of the total column amount of CO₂, CH₄ and CO from two mobile platforms located inside and outside the city plume (usually at upwind and downwind locations on the opposite sides of the city of St. Petersburg) combined with the *mobile circular measurements of tropospheric column amount of NO₂ from the third mobile platform moving in a non-stop mode, the latter measurements are used for the real-time control of the megacity plume evolution*" (beginning of Section 2, orig. version). Generally, the DOAS measurements confirmed the HYSPLIT forecast. However, on one day of experiment this was not the case, and the FTIR measurements location was timely corrected according to the data of DOAS observations. This is mentioned at the end of Section 3.1, lines 217-221, orig. version.

The referee is right, the methodology of mass balance approach was applied to estimate NO_x flux in exactly the same way as it was done for all other species (CO₂, CH₄ and CO). We do mention this in the manuscript: "The summary of the EMME-2019 results and the comparison with the flux estimates for St. Petersburg based on in situ measurements, as well as independent literature data, are presented in Table 4 (orig. version) for CO₂, CH₄, CO and NO_x (*the latter were derived from mobile DOAS measurements of tropospheric NO₂ in the vicinity of upwind and downwind FTIR observations*)" (line 401-404 of Section 5.1, orig. version). Indeed, much more data of NO₂ measurements is available from our circular DOAS observations, but its interpretation is a subject of separate study and is beyond the scope of the manuscript under review. Finally, an answer to another referee's question here: no, we did not use the NO₂ data to correct the FTIR measured data.

The treatment of uncertainties is all based on the obtained/measured variability of the parameters used to calculate the flux (total column, effective path and wind).

In my mind this is an assessment of the random uncertainty. However there is no mentioning of systematic errors of any of these parameters. Please add a discussion about this and change absolute uncertainties to random uncertainty.

The following discussion was added in the paper:

To evaluate systematic error of the area flux (δF_{sys}) we should first estimate the systematic errors δL_{sys} , δV_{sys} and $\delta \Delta TC_{sys}$ of corresponding parameters L , V and ΔTC in Eq.2. In contrast to δL_{sys} and δV_{sys} , the contribution of systematic component of $\delta \Delta TC_{sys}$ into δF_{sys} is negligible. This is due to the high accuracy of the COCCON observations of gas columns which are calibrated against WMO scale. In Eq. 2 we use an assumption that an air parcel moves along a straight line but obviously this is not true. For the whole ensemble of HYSPLIT trajectories simulated for all days of the city campaign we calculated the maximum relative difference between the true lengths of HYSPLIT trajectories and our straight line approximations of L . This value equals to ~4% which is considered as an estimation of the relative systematic error δL_{sys} . According to the information on wind speed observed during the field campaign (see Appendix A, Table A3), the mean relative difference between HYSPLIT and GDAS data on wind speed is of 14±22%. Hence, the estimation of the systematic error of area flux δF_{sys} due to the systematic errors of all parameters in Eq.2 gives the value 18%.

In the CO₂ and CO data there is a factor of two difference between the column measured data and the one measured by in situ data. This is explained by the fact that the CO₂ and CO emissions are released from high chimneys (200m). However the mixing layer should be several hundred meters (at minimum) at solar conditions and the pollutants should therefore well mixed at some distances from the chimney (>1 km). This was also supported by kite measurements. In addition a considerable portion of the CO₂ should come from transport sector. The discussion should be improved on this topic.

We agree with the referee that this issue requires some more discussion. Taking into account that this topic is specific, we put the extended discussion in Appendix C:

Appendix C: Comments on transport of the pollutants from elevated sources

We illustrate transport of the pollutants from elevated sources with a HYSPLIT simulation (see Fig. C1). We selected one of the days of EMME (April 16, 2019) and simulated the CO₂ emission from a 180-meter chimney of the thermal power station mentioned above in the main text of the article. The plot presents a 34-hour trajectory of the mass-weighted CO₂ plume position (the centroid of the plume) on the geographical map (top panel) and using the altitude scale (bottom panel). One can see that the plume centroid starts its movement from the chimney location at ~180 m altitude (12:00 of April 15) and raises up to ~500 m in one hour; then it does not fall below the level of ~350 m during its "flight" length of more than 300 km. The detailed analysis of respective vertical profiles of CO₂ concentration shows its maximum at ~500 m, being 1.2 times higher than that on the surface at start and 3.6 times higher than that on the surface at the end of the plume trajectory. Thus, the probability to register high concentrations corresponding to the centroid of the plume by surface-based observations can be estimated as very low. Moreover, polluted air mass from a chimney is more likely to rise up, rather than descend to the ground due to two reasons: (1) the vertical velocity of the air pollution jet emitted from a chimney can be rather high; (2) the temperature of a plume released from the chimney is usually significantly higher than the temperature of the ambient air causing the buoyancy effect.

Elevated air sampling using kite launches was performed only twice during the EMME campaign, therefore the results of these kind of measurements could not be considered as a reliable confirmation of the absence of elevated plumes. The presence of the elevated plumes of CO and CO₂ could be also confirmed by the following evidence. The comparison of the values of area fluxes (F , see Table 1) estimated using in-situ measurements (column #4) and FTIR observations (column #2 and #3) shows that for CH₄ which sources are mainly located on the ground surface we obtain significantly lower difference in corresponding F values than for CO and CO₂.

NOAA HYSPLIT MODEL (mass-weighted centroid position of CO₂ plume)
 Forward trajectory starting at 1200 UTC 15 Apr 19
 GFSG Meteorological Data

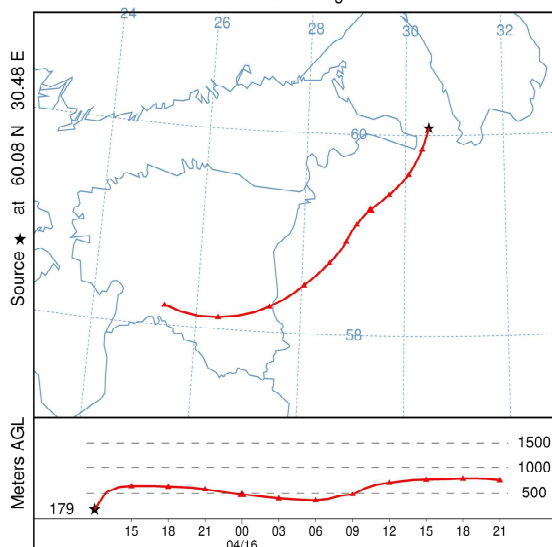


Figure C1: Evolution of the mass-weighted centroid position of the CO₂ plume taken as an example (see text).

Specific comments

P3: Row 83: When making reference to other studies it would be relevant to add similar large scale measurements by mobile FTIR (Solar Occultation Flux technique) and mobile DOAS which has been applied for large scale flux measurements for at least a decade by now : e.g. 1. de Foy, et al., (2007) Modelling constraints on the emission inventory and on vertical dispersion for CO and SO₂ in the Mexico City Metropolitan Area using Solar FTIR and zenith sky UV spectroscopy. *Atmospheric Chemistry And Physics* 7, pp. 781-801. DOI: 10.5194/acp-7-781-2007. 2. Mellqvist, et al., (2010) Measurements of industrial emissions of alkenes in Texas using the solar occultation flux method. *Journal of Geophysical Research - Atmospheres* 115. DOI: 10.1029/2008JD011682. 3. Johansson, J., et al. (2014) Emission measurements of alkenes, alkanes, SO₂, and NO₂ from stationary sources in Southeast Texas over a 5 year period using SOF and mobile DOAS. *Journal of Geophysical Research-Atmospheres* 119, no. 4, pp. 1973-1991. DOI: 10.1002/2013jd020485. 4. Johansson, et al. (2014) Quantitative measurements and modeling of industrial formaldehyde emissions in the Greater Houston area during campaigns in 2009 and 2011. *Journal of Geophysical Research-Atmospheres* 119, no. 7, pp. 4303-4322. DOI:10.1002/2013JD020159. 5. Kille N, et al, The CU Mobile Solar Occultation Flux instrument, *AMT*, 10, 373-392, 2017

The following text has been added in the introduction section:

... 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. Vogel et al. (2019) investigated the Paris megacity emissions of CO₂ by

coupling the COCCON observations and atmospheric transport model framework (CHIMERE-CAMS) simulations.”

.....

“... De Foy et al. (2007), Mellqvist et al. (2010), Johansson et al. (2014), and Kille et al. (2017) have applied mobile FTIR (Solar Occultation Flux technique) and mobile DOAS techniques to the large scale flux measurements.

P 5, row 121: You claim that the DOAS measures tropospheric columns. Please elaborate in a few sentences what is actually measured, even though you refer to previous studies. Are you using multiaxis measurements to derive absolute columns or is it differential columns assuming that the upwind measurements is free from tropospheric NO₂, and hence that the differential measurements corresponds to the tropospheric absolute column.

In the revised version of our manuscript we added a text with some more details of our DOAS measurements (see above). We are not using multiaxis (or MAX-DOAS) observations. Our DOAS measurements are just zenith-sky, and we specify that in the manuscript.

P5, row 132. Add references from other places on mobile DOAS, e.g. Johansson, M et al., Mobile mini-DOAS measurement of the outflow of NO₂ and HCHO from Mexico city, ACP, 9(15):5647-5653, 2009. Rivera, C. et al., (2010) Quantification of NO₂ and SO₂ emissions from the Houston Ship Channel and Texas City industrial areas during the 2006 Texas Air Quality Study. Journal of Geophysical Research - Atmospheres 115. DOI: 10.1029/2009JD012675.

In the revised version we added the following sentence and significantly expanded the list of relevant references:

In general, such observations have been proved to be an efficient technique to derive the anthropogenic NO_x flux in many studies worldwide (see e.g., Johansson et al., 2008, Rivera et al., 2009, Johansson et al., 2009, Rivera et al., 2010, Ibrahim et al., 2010, Shaiganfar et al., 2011, Wang et al., 2012, Shaiganfar et al., 2015, Wu et al., 2017, Shaiganfar et al., 2017).

P6, row 171: This sentence is unclear rewrite it. For instance Table 1 presents daily information ...

In the revised version we added the following text:

Table A1 (see Appendix A) presents daily information on the location of FTIR spectrometers during the campaign, FTIR spectrometer identifier, number of bags of air samples, flight of a kite and air sampling altitude.

P8, row 128: Define X_{gas} (is it against pressure?) and motivate why you introduce this. Would it not be more appropriate to compare total columns instead of X_{gas} since TC is the ones used for the flux.

For the cross-calibration of the EM27/SUN spectrometers we used XCO₂, XCH₄, and XCO values as strongly recommended in the special study by Frey et al. (2015). To define X_{gas}, we added the following text:

The ratio of the target gas TC to the retrieved O₂ TC which is suggested to be known and constant, gives us the column-averaged dry-air mole fraction (X_{gas}) of the target gas (Wunch et al., 2011; Frey et al., 2015):

$$X_{gas} = 0.2095 \frac{TC_{gas}}{TC_{O_2}} = \frac{TC_{gas}}{TC_{dry\ air}}, \quad (1)$$

where X_{gas} - column-averaged dry-air mole fraction of the target gas (unit: dimensionless quantity), TC_{gas} - total column of the target gas (unit: molec. m⁻²), TC_{O_2} - total column of O₂ (unit: molec. m⁻²), $TC_{dry\ air}$ - dry air total column (unit: molec. m⁻²). Using X_{gas} helps to reduce the effect of various possible systematic errors (Wunch et al., 2011). To provide the compatibility of EM27/SUN measurements to WMO scale and for consistency reasons, the retrieval software used for processing the EM27/SUN spectra also performs a post-processing (Frey et al., 2015). Finally, we had at our disposal both the TC_{gas} and X_{gas} for each day of measurements at each observational location.

P8, row 232: The comparisons between the two spectrometers is very convincing. Nevertheless, it only shows how the spectral properties of two spectrometers influences the statistical error of the measurements. Please comment how this information was used.

After cross-comparison procedure we used obtained regression parameters to scale the data. The result after the scaling process is shown in Figure 5. We explain it in the revised version:

The calibration factors obtained as a result of side-by-side comparison were used to convert XCO₂, XCH₄, and XCO measured by spectrometer #80 to the scale of spectrometer #84. The results of cross-calibration help to avoid an additional source of systematic error in the estimation of area fluxes.

P 9, 244: I think this section should be more detailed wrt the spectroscopy. At least a couple of general sentences for how te retrieval is done and if there are interfering species etc could be helpful,

In the revised version we added the following text in section **4.1 FTIR and DOAS data processing**:

...For the retrievals of the total columns of O₂, CO₂, CO, H₂O, and CH₄, the spectral regions recommended by Frey et al. (2019) and Hase et al. (2016) were taken. We present these intervals in the respective order: 7765 – 8005 cm⁻¹ (the main interfering gases are H₂O, HF, CO₂), 6173 – 6390 cm⁻¹ (the main interfering gases are H₂O, HDO, CH₄), 4210 – 4320 cm⁻¹ (the main interfering gases are H₂O, HDO, CH₄), 8353 – 8463 cm⁻¹, and 5897 – 6145 cm⁻¹ (the main interfering gases are H₂O, HDO, CO₂). The EM27/SUN spectrometer has low spectral resolution of 0.5 cm⁻¹. Therefore the TCs are derived from the FTIR spectra by scaling of a priori profiles of target gases (Frey et al., 2019).

Special note:

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