Responses to Referee 3:

Thank you very much for your significant and useful comments on the paper "O_2:CO_2 exchange ratio for net turbulent flux observed in an Urban Area of Tokyo, Japan and its application to an evaluation of anthropogenic CO_2 emissions" by Ishidoya et al. The title of the paper has been changed from the ACPD paper. We have revised the manuscript, considering your comments and suggestions. Details of our revision are as follows;

Main concern:

My main concern is the lack of met-related filtering of the atmospheric CO_2 and O_2 data prior to deriving the fluxes. I feel that the data handling as it is currently presented is perhaps too simplistic and should be taken further. I would like to see: a) filtering of the data to exclude periods that are highly influenced by regional not local fluxes (i.e. using associated met data, other tracers, or the concentration measurements themselves); b) more robust quantification of the ORs. While I can see the authors have attempted some robustness by calculating the ORs over two different time horizons (1-day and 1-week), I think this approach is not the best. Usually ORs are most robust during the onset of an atmospheric 'event' but not during the recovery phase when atmospheric conditions are unstable. So I would recommend only calculating ORs during the onset of atmospheric events. Also, a more robust approach to calculating ORs might consider other factors such as wind direction. This might also yield a more in-depth analysis of the OR results. I would also caution the authors about ascribing variations they see in the atmospheric ORs to changes in local fluxes, unless they can discount the influence of seasonal/diurnal atmospheric dynamic effects.

Considering your comments, we have added the discussion including filtering of the data using wind direction (line 191-210 and Fig. 6). For the analyses of specific events, we have reported the OR values and simultaneously-measured $PM_{2.5}$ aerosol composition for a week-long pollution event by Kaneyasu et al. (2020) (line 208-210). Considering the results of the discussion, we decide to use all the O_2 and CO_2 concentration data without filtering by the wind direction, to increase the number of data

points for calculating OR_F and OR_{atm}; this is consistent with the purpose of this study to derive representative OR values at the YYG site in order to validate the CO_2 emission inventory updated from Hirano et al. (2015). It is noted that we have newly added discussion to estimate the average diurnal cycles of CO_2 fluxes from gas and liquid fuels consumption separately by using the OR_F, CO_2 flux, and inventory-based CO_2 emission from human respiration, in order to validate the inventory-based CO_2 emissions from gas consumption and traffic (line 290-344, Fig. 9 and Fig. 10).

Specific comments:

1) Several times in the introduction, the authors mention that ORs can be used to separate out the contribution of different sources to the observed CO_2 flux. I cannot think of a way this would work in reality without additional information (i.e. from bottom up inventories) unless one has a very idealised case with discrete sources coming from very different wind directions, for example. But for most cities, the sources are mixed. Ultimately, the measured OR will be a mixture of all the sources in the footprint, so it could be used to 'check' modelled OR estimates (although two 'wrongs' can also make a 'right'), but it cannot be used in itself to distinguish CO_2 fluxes from different sources.

We agree with you that OR can be used to check modelled OR but cannot be used in itself to distinguish CO_2 fluxes from different sources. Therefore, we have changed some sentences, e.g. from "…then the information can be used to separate out the contributions of the gaseous, liquid, and solid fuels, and the terrestrial biospheric activities to the observed CO_2 flux" to "…then such information can be used as a useful constraint for evaluating the contributions of the gaseous, liquid, and solid fuels, and the terrestrial biospheric activities to the observed CO_2 flux" (line 44-46). Moreover, as mentioned above, we have newly added discussion to estimate the average diurnal cycles of CO_2 fluxes from gas and liquid fuels consumption separately by using the OR_F, CO_2 flux, and inventory-based CO_2 emission from human respiration, in order to validate the inventory-based CO_2 emissions from gas consumption and traffic (line 290-344, Fig. 9 and Fig. 10).

Several times, the flux footprint vegetated area is stated at 9% in summer and 2% in winter. The authors should state how these values are derived. They also seem too low, based on the images given in figure 1. If there is a strong seasonal difference in the footprint of the measurements between summer and winter, how are the authors sure that the OR results they obtain are related to changing flux patterns/behaviour and not simply caused by the changing footprint?

The vegetation area was calculated from the area included in the aerial photo in Fig. 1 by considering the contribution to flux. The calculation method for the footprint is based on the model of Neftel et al. (2008) (line 63). It is noted the footprint and the caption of Fig. 1 have been revised. As you pointed out, there is a seasonal difference in the footprint between summer and winter due to the seasonal difference of the prevailing direction of wind. However, as shown in the contour lines in Fig. 1, which indicate contribution in measured flux (60, 50, 40, 30, 20 and 10% from outside to inside), the dominant contribution to flux is from the adjacent area of the observation tower; within about 300 m from the tower in both seasons. Land cover is nearly uniform in this dominant footprint area as shown in Fig.1. Therefore, the observed OR_F values are determined mainly by the O_2 and CO_2 fluxes from the urban area and the effect of seasonal difference in the footprint to the OR_F would be relatively small.

2) Lines 73-74: How did the authors subsample from such a high flowrate without using a tee and causing fractionation of O_2 wrt N_2? Please clarify, since this is an important technical point.

Lines 80-87: We have added the sentences to show the subsampling method and discuss the possible fractionation for the measurements in this study.

3) Lines 91-96: I'm not sure that the logic is valid here. Since ORs are calculated from regressing two sets of data, they are most sensitive to inaccuracies at the high/low ends of the scale. So I think the authors might find the uncertainties in OR are larger than the 1% uncertainties at the high end of the CO_2 scale. The easiest way to check is to recalculate some ORs using a 1% difference in the high CO_2 values and see how large the difference in OR is. My guess would be it's more like 10%.

Lines 101-108: The OR is calculated as a ratio of difference of O_2 concentration to that of CO_2 concentration, so that we consider the effect of the span-difference of CO_2 on

the OR does not depend on the absolute value of the CO_2 concentration, as following idealized tables:

O_2 anomaly High (ppm)	O_2 anomaly Low (ppm)	CO_2 on scale-1 High (ppm)	CO_2 on scale-1 Low (ppm)	OR
-400	-600	600	500	2
-400	-600	400	300	2

O_2 anomaly High (ppm)	O_2 anomaly Low (ppm)	CO_2 on scale-2 High (ppm)	CO_2 on scale-2 Low (ppm)	OR
-400	-600	612	510	1.96
-400	-600	408	306	1.96

*OR values are calculated by "–(O_2_high – O_2_low)/(CO_2_high – CO_2_low)", and the span-difference of CO_2 between scale-1 and scale-2 is 2%.

We have also modified the sentences and allowed the uncertainty of within 3% for OR, which is larger than the ACPD, due to the span-uncertainties of O_2 and CO_2 concentrations.

4) Lines 115-116: two things here. Firstly, I would caution against attributing changes in the atmospheric data to changes in fuel usage without very strong evidence, ideally from multiple sources. Such changes can sometimes be caused by seasonal changes in atmospheric dynamics or changing footprint, see my comment above. Secondly, winter is usually associated with more boundary layer turbulence, not more stratification. If the authors disagree, please can they provide a citation to back up this statement, which seems to me to be erroneous.

As already mentioned above, we have newly added discussion using the CO_2 emission inventory data of gas consumption, traffic and human respiration around YYG to show the evidence for changes in fuel usage (line 290-344, Fig. 9 and Fig. 10). The sentences and figures have been added to show the evidence that O_2 is consumed within the urban canopy at YYG especially in winter (line 127-132 and Fig. 3), and the words "a more stable stratification of surface atmosphere" have been changed to "a temperature

inversion near the surface" to make the meaning clearer. It should be note here that the stable layer can be found mainly in winter (Kanda et al., 2005), meaning less turbulence in winter than in summer.

M. Kanda, R. Moriwaki and Y. Kimoto, Temperature Profiles Within and Above an Urban Canopy, Boundary-Layer Meteorology volume 115, 499–506, 2005.

5) Lines 177-178: I think the authors state here that there was no coal fluxes observed because no ORs were 1.17? If so, I would strongly advise the authors retract this statement, since it is very possible that a mixture of coal and gas could give a ratio that looks like liquid fuel, and yet perhaps there was no liquid fuel burnt at the time. If there is independent evidence for expecting no or very little coal (such as from an inventory) then please provide this here.

Lines 221-225: The sentences have been added to show independent evidence for expecting very small contributions of coal.

6) Line 194: "on the other hand" used twice in same paragraph. Suggest to rewrite one of them. Or better still to omit entirely, since this is rather colloquial language for such a publication.

Lines 239-242: The words "on the other hand" were removed, as suggested.

7) Line 202: Suggest to rewrite "seasonal "climatological" diurnal cycles" as I am not sure what the authors mean. I think what is meant is the average diurnal cycle in different seasons.

Line 254: The words "seasonal "climatological" diurnal cycles" have been changed to "average diurnal cycles".

8) Lines 219, 221: I would advise caution again here, unless there is independent evidence to back these statements up. It would also be nice to see how much diurnal variation there is in the site footprint, in addition to the seasonal variation.

As noted in our response to your comment No.1, the flux footprint was mainly located around the tower. The footprint had diurnal variation in its location, however it was still located in the relatively homogeneous area around the tower.

Considering your comments, we made some revision in our manuscript (lines 271-273, 290-344, Fig. 9 and Fig. 10). We have modified the sentences considering your comments, and we have added sentences and figures to discuss the estimations of the average diurnal cycles of CO_2 fluxes from gas and liquid fuels consumption separately by using the OR_F, CO_2 flux, and inventory-based CO_2 emission from human respiration, in order to validate the inventory-based CO_2 emissions from gas consumption and traffic. The inventory-based emission data have been updated from Hirano et al. (2015) for the present study. We hope these revisions will meet your suggestion to show independent evidence to back the statements up.

9) Lines 237-247 and corresponding text in conclusions: I do not see the value in this paragraph or it's relevance to the rest of the paper. The authors state that the O_2 urban fluxes are very large compared to the global mean O_2 fluxes, but the global O_2 decrease accounts for O_2 fluxes from all urban regions, so I'm not sure what the point of the comparison is. And as the authors themselves state, it is unrealistic that urban O_2 depletion would lead to atmospheric O_2 falling to levels that are dangerous for human health (perhaps this is possible for isolated indoor environments, but not in the free atmosphere – this has been debunked many times now by many people). I would recommend the authors remove this paragraph and focus solely on the OR analyses.

Lines 284-289: We agree with you that the statements in the paragraph do not have enough value to discuss in detail. Therefore, the sentences have been much shortened and started the phrase "In this regard…" to clarify that is just for reference, and we have focused on the OR analyses combined with the inventory-based CO_2 emissions prepared for the present study (line 290-344, Fig. 9 and Fig. 10).

10) Figure 2: It is hard to see the seasonal difference of delta O_2 and delta CO_2 with the current y-axis scaling.

Figure 3: We have added the figure to show the O_2 and CO_2 concentrations, ΔO_2 and ΔCO_2 for the period December 16 – 23 and July 1 – 9, 2016, to see the seasonal difference.

11) Figure 3: please separate the O_2 and CO_2 grey data points with more white space so the two time series datasets can be viewed independently/more easily.

Figure 4: The figure has been modified, as suggested. It is noted the number of the figure has been changed from that in the ACPD paper.

12) Figure 4: do these regression fits account for the difference in measurement precision between CO_2 and O_2? Also, please state whether the fits account for both x and y uncertainties.

Lines 170-178: We have changed the regression method to Deming regression throughout the paper for calculating OR, in order to account not only for the difference in measurement precision between CO_2 and O_2 but also for both x and y uncertainties.

13) Figure 6: what are the open circles? The evening peak seems to occur too late in the day to be accounted for by traffic alone (especially in winter). Also, this peak is much broader than the morning peak, suggesting there is a net flux of traffic out of the region over time (whereas presumably this is not the case). I think some more in-depth analysis into these patterns would be useful here.

Figure 8: The words to explain the filled and open circles in the upper panel have been added to the figure caption. It is noted the number of the figure has been changed from that in the ACPD paper. As to the detail analyses of the morning and evening peaks, we have added the OR analyses combined with the inventory-based CO_2 emissions as mentioned above (line 290-344, Fig. 9 and Fig. 10).