Reply to Reviewer 2

We sincerely thank the reviewer for carefully reading our manuscript and for providing valuable and constructive comments and suggestions, all of which we have accepted. Below, we present our point-by-point responses to each comment.

This manuscript presents simultaneous measurements of NO2 by ASKY and near-surface CO2 concentrations using LI-7810 and G4301 trace gas analyzers. [ΔCO2] was also estimated from the positive correlation of CO2 to NO2 and BC, respectively. This work is scientifically meaningful and contains annual measurement data, but need more clarification to be published at AMT.
To clarify the usefulness of simultaneous near-surface CO2 and NO2 measurements for simple and accurate monitoring of fossil-fuel combustion-derived CO2 in the Tokyo megacity, thorough analysis is required with appropriate literature surveys.

Reply: We appreciate your positive assessment.

To estimate CO2 concentrations from NO2 and/or BC using their relationship, it is assumed that the NO2/BC emission sources are related to that of CO2. The relationship depends on region, season, time scale, meteorology, winds in particular, lifetime in the atmosphere, the ratio between NO and NO2 etc. High correlation between the two species does not guarantee the estimation of one species' concentrations, which may lead biases. Comparison of measurements from the remote sensing instrument, MAX-DOAS to in situ instrument may represent spatial scale. At the same time, this assumption depends on the carbon dependence of energy source and emission reduction mechanism from region to region. Japan is one of the leading countries in minimizing NOx emissions and utilizing renewable energy source with high technologies. Thus, it requires more thorough evaluation process to infer CO2 concentration from NO2.

Reply: We understand your comment as pointing out that estimating CO_2 concentrations based on the correlation between CO_2 and NO_2 (or BC) and is problematic. We agree that such an estimation method would indeed be inappropriate. Therefore, we would appreciate it if you could understand that our study does not employ such an approach. In our study, we defined the CO_2 concentration observed when NO_2 levels were extremely low as the baseline (denoted as $[CO_2^*]_N$). In other words, NO_2 was used only to identify the CO_2 data corresponding to periods when NO_2 concentrations were very low. Because this identification was made using the 5th-percentile value, our approach does not depend on whether Japan had advanced technologies for reducing NO_x emissions. As you pointed out, increases above the baseline can be influenced by various factors. Keeping this in mind, because the increase ($[\Delta CO_2]_N$) was found to correlate well with NO_2 concentrations, we argue that the primary driver of this increase is fossil fuel combustion. We also agree with your comment that the correlation between the CO_2 enhancement and NO_2, as well as their ratio, can depend on meteorological conditions, photochemical processes, and other factors. For this reason, we emphasize that we did not estimate fossil fuel CO_2 concentrations from the correlation. We have revised the relevant section of the manuscript to make this clearer to readers.

LI 7810 trace gas analyzer measures CH4, CO2 and H2O at wavelengths of 1.6 micron, where CO2 absorption signal is weak. Thus,
Authors need to elaborate further on Section 4 (Conclusion) by including quantitative evaluation results and relevant statistical metrics to support the main findings.

Reply: In response to the comment, quantitative evaluation results and relevant statistical metrics to support the main findings are now included in Section 4 (Conclusion) of the revised manuscript.

94: The use of 70 degree instead of 90 degree was justified with the signal stabilization. Can you suggest reference or supporting evidence?

Reply: Yes. Instead of 90°, a 70° elevation angle was adopted as the reference in order to reduce the variation range of signals measured at all elevation angles, while keeping the integration time constant. In the vertical profile retrieval, the elevation angle settings were fully accounted in the radiative transfer calculations used to compute the differential air mass factors (e.g., Irie et al., 2011, 2015, 2019). These points are now mentioned in the revised manuscript, and the corresponding references have been added.

Fig. 5: There are clear two data groups above and below the linear regression line. Would it be possible to find the cause of this difference ?

Reply: In response to the comment, we performed an additional analysis for Fig. 5, which now reveals temporal variations on a monthly time scale. We found that the two groups appeared in different seasons: one corresponding to the period from April to August when H2O and temperature increase, and another corresponding to the period from September to December when they decrease. Comparing these groups, the slopes of the regression lines were nearly the same, but the intercepts differed. This difference appears to be related to temperature and/or LI-7810 instrumental drift, but we understand that further analysis using multi-year data is needed to identify the cause.