Reviewer #2:

This study presents a new top-down superposition column model to estimate daily NOx and CO2 emissions from the largest city Wuhan in central China. It gives a very detailed description of this model, and the application to Wuhan clearly demonstrates the promising future. Overall, I think this manuscript is well structured, and the topic is suitable for ACP.

Major comments.

I think the authors need to clearly describe their way of calculating the uncertainty of their estimates. According to Section 2.6, large uncertainties are attached to the parameters used by the model (20% for OH, 10% of NOx/NO2, 30% for CO2-to-NOx, 15% of NOx lifetime). But the reported uncertainty from the text and Figure 3 is much lower (probably less than 10%). Could the authors specify how they calculate the full uncertainty? Have they used a Monte Carlo (or a similar) method to account for the uncertainty of each sub-process together?

Response: Thank you for this suggestion. We ran a test by randomly choosing parameter values within their uncertainty ranges for 20 times to predict an ensemble of NO_x and CO_2 emission outcomes. Then the ratio of the standard deviation to the mean value of the 20 emission outcomes is regarded as the uncertainty on NO_x and CO_2 emission caused by uncertainties in the corresponding parameters. The results in terms of uncertainties on NO_x and CO_2 emissions are listed below:

factor	**uncertainty**	**Influence on NO_x and (or) CO_2 emissions**
Satellite NO_2 retrieval	**±20 %**	**20 %**
OH concentration	**±20 %**	**3 %**
NO_2/NO_x ratio	**±10 %**	**8 %**
Wind field	**±20 %**	**17 %**
CO_2-to-NO_x emission ratio	**±30 %**	**30 %**

The areas of the study domain may also lead to uncertainty in NO_x and CO_2 emissions. We have narrowed down our study domain to check the sensitivity of our method to the chosen study domain (see Fig. S7). In the test, the study domain is narrowed down to 84

km in diameter, and, as expected, the result turns out to be structurally different from that with the 186 km diameter domain, for the mean OH concentration is lower in the city center, leading to longer fitted NO_x lifetime. However, the change in fitted NO_x lifetime and NO_x emission is less than ±15%. So we give an 15% uncertainty in NO_x emission estimation caused by the size in the area of the study domain. All the uncertainty factors and their influence on NO_x and CO_2 emission estimation are listed in Table S2.

Finally, considering that all these parameters are independent from each other, we use the root mean square sum of the contributions to represent the overall uncertainty estimation, which we quantify for NO_x emission on a single day at ~31 %, and for CO_2 emission at ~43 %. We updated our revised manuscript accordingly on Page 7-8, Line 192-202, Fig. 3 and Fig. 6 in the revised manuscript and Table S2 in the revised supplementary material.

Minor comments.

Line 21. Please specify the uncertainty. I believe the uncertainty of bottom-up inventories should be much larger than 3%.

Response: We agree. Differences between emissions from two inventories for the year 2019 were <3%, but the uncertainty is arguably larger than that. We rephrased the sentences: 'We estimated daily NO_x and CO_2 emissions from Wuhan between September 2019 to October 2020 with uncertainties of 31% and 43% respectively. Our estimated NO_x and CO_2 emissions are verified against bottom-up inventories with small mutual deviations (< 3 % for 2019 mean, ranging from -20 % to 48 % on a daily basis).'

Line 77. The retrieval methods can considerably affect the column concentrations. So it is better if the authors can comment on the related effects if using a newer version of TROPOMI data.

Response: This is exactly why we used two contemporary versions of the operational TROPOMI NO_2 products (v1.3 and v2.3.1) and evaluated the impact of the retrieval version on our emission estimates. Improved (residual) cloud pressures correct the low bias of v1.3 data compared to OMI and ground-based measurements over east China

(Wang et al., 2020; Liu et al., 2020). In addition, an improved treatment for the surface albedo increases the columns for cloud-free scenes (Van Geffen et al., 2022). Compared to the earlier version, the v2.3.1 dataset has 10–40 % higher tropospheric NO_2 columns over polluted scenes due to the improved cloud retrieval and other algorithm updates (Van Geffen et al., 2020; Riess et al., 2022). Over Wuhan we find an average increase in tropospheric NO_2 of about 25%, but there are also differences between the two versions in terms of spatial and temporal distribution (Fig. S2). According to Fig. S2, the increase in v2.3.1 is much stronger over a polluted area (city center) and polluted period (9 September and 3 October 2019). The estimated NO_x lifetime and emissions from the two datasets for the whole study period are presented in Fig. S5. On average, using the TROPOMI-v1.3 data leads to 13% lower NO_x emissions from Wuhan than using the TROPOMI-v2.3.1 data. The NO_x lifetime estimated from TROPOMI-v1.3 data is 5% shorter than that from TROPOMI-v2.3.1, which may be attributed to the fact that the TROPOMI-v2.3.1 data has a higher ratio between city center to the background. This information is added in the revised manuscript in Page 3 Line 79-91 and Page 9 Line226-231, and the revised Fig. S6.

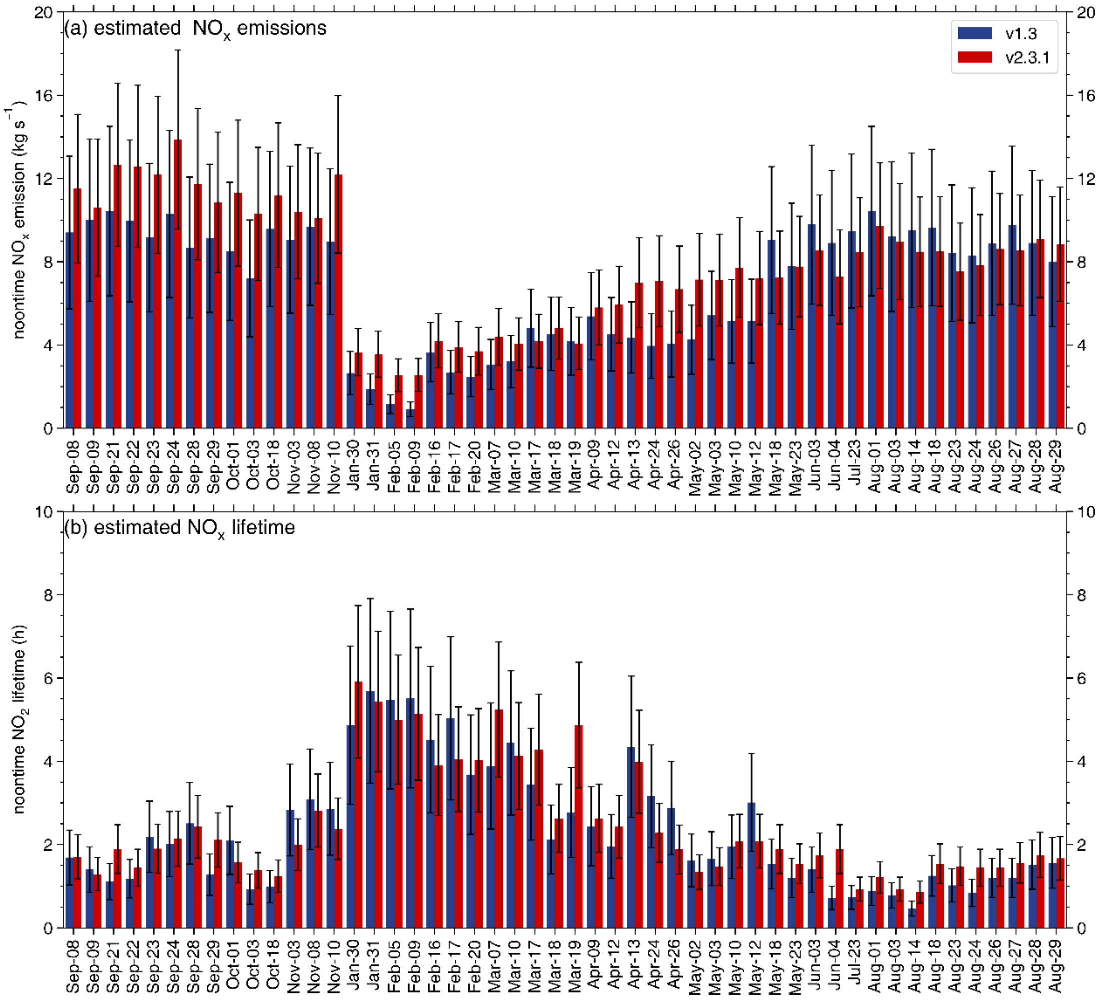

Figure S6. Estimated NO$_x$ (a) emissions and (b) lifetime over Wuhan during the study period based on the TROPOMI-v1.3 (blue bars) and TROPOMI-v2.3.1 (red bars) datasets. The error bars denote the corresponding uncertainty.

Line 88. Area-weighted sampling?

Response: The satellite data is sampled in the regular longitude-latitude grid at a resolution of 0.05° (lon) × 0.05° (lat), and it is approximately 6 km × 6 km in the area of each grid. We didn't use an area-weighted sampling method, and a single pixel is only considered when its center falls in the cell. The NO$_2$ column density in each grid is calculated as the arithmetic mean of all the considered pixels.

Line 105. Please specify the version of GEOS-Chem and give the reference. Also, give the full names of ERA5 and ECMWF

Response: added. We use the version 12.1 of GEOS-Chem model with a horizontal resolution of 0.25° × 0.3125° (~ 30 × 37.5 km^2) to provide the a priori guesses for chemical parameters relevant to daytime NO_x. The wind field is from ERA5 (ECMWF Reanalysis v5), the fifth generation ECMWF (European Centre for Medium-Range Weather Forecasts) atmospheric reanalysis of the global climate.

Line 148. The correlation coefficient of pixels along the wind direction is not a very useful metric here because these pixels are not independent of each other. It is better to show the mean bias along with the correlation coefficients.

Response: The reviewer's point is well-taken. The mean bias is now also included in the revised manuscript in Page 6 Line 158-161, and is displayed in the revised Fig. 1. Both the correlation coefficient and the mean bias are useful metrics to evaluate the performance of the superposition model. The correlation coefficient quantifies the success with which the fitting model reproduces the observed line densities, and the mean bias describes the deviation of fitted line densities from the observations. Compared to fitting results with a constant background value, we obtain a better correlation (up to 25%) and lower bias (nearly 50% lower) between fitted and observed NO_2 line densities then when fitting with a linearly changing background value.

Line 342. Please specify the resolutions here.

Response: This sentence is rephrased as "Compared to previous studies, our work shows that satellite measurements can provide detailed information on sub-city scale NO_x and CO_2 emissions on daily basis".

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Wang, C., Wang, T., Wang, P., and Rakitin, V.: Comparison and Validation of TROPOMI and OMI NO2 Observations over China, Atmosphere, 11, 10.3390/atmos11060636, 2020.