Dear Reviewer,

We really appreciate your careful review and insightful comments that helped a lot to improve the analysis and writing of the manuscript. the point-by-point response to your comments is listed below and the revisions/additions/edits are shown in the tracked-change file.

The manuscript presents an interesting investigation using TROPOMI NO2 column data in combined with superposition column model to estimate the emission and lifetime of NOx. Specifically, the study focuses on the derive the NOx emissions and lifetime over Wuhan for 335 clear sky days between May 2018 and December 2023, with the variability of emissions being evaluated to investigate the effectiveness of the emission control strategy. There are some interesting findings resulted from the study. However, the reviewer has some concerns about the novelty of the methods and the significance of the results. See detailed comments below.

Major comments:

1. This paper looks like an extension of the authors' ACP paper published in 2023. Similar methods are applied to the TROPOMI data (with version change though) over the same region, and the main difference is that this study extends the study period from 2019-2020 to 2019-2023. Because of the overlap with the authors' previous study, the reviewer is concerned about the novelty of this manuscript, especially since the technical approach has been proposed in their 2023 paper. The authors should clarify in the introduction how this manuscript differs from the previous study, and what would be the novelty of this study.

Response: Thank you for the comment. In the revision of the work, we made substantial modification to the superposition column model. We discard the GEOS-Chem simulated OH concentration in the estimation of NO_2 chemical loss rate to get rid of the dependence on CTMs and reduce computational burden, and also avoid the uncertainty induced by the OH concentration. We have added this information in Page 3, Line 75-78 in the revised manuscript. This work is not just an extension in study period of the previous work, we have thoroughly discussed the uncertainty and limitations of the superposition column model on every aspect, providing a reference for future studies to use satellite data to constrain NOx emissions.

2. It's unclear how the NOx lifetime is calculated in GEOS-Chem. The model approach gives an effective lifetime of the entire plume, but the actual chemical lifetime can vary from source to downwind. The effective lifetime can be further confounded by mixing of plumes from multiple directions. I'd suggest the authors clarify the meaning of lifetime in the manuscript, and the limitations of using the model approach to estimate NOx lifetime.

Response: The reviewer's comment is taken and we have made it clear in the revised manuscript that the 'lifetime' mentioned in this work is the 'chemical lifetime' of NO_x, and yes the method gives mean chemical life of the entire study domain. The chemical lifetime explains only a part of NO_x loss in the atmosphere, and NO_x chemical lifetime estimated from this method is found to decrease when wind speed increases, which is caused by the stronger ventilation of NO_x. We have discussed this in Page 14-15, Line 354-368 in the revised manuscript.

3. The authors showed strong dependence of the emissions and lifetimes on wind field, which does not necessarily mean the NOx emissions vary with wind, but rather due to the limitation of the model and the way the model defines background NO2. This is not a scientific finding, so I think it's better to be included in the uncertainty discussion

Response: We agree with the reviewer's point that the dependency of the estimation on the wind field reveals the uncertainty of the method.

The lower estimation of NO_x emissions under easterly winds indicates that the method underestimates NO_x emissions when there is NO_2 hot spots in the upwind region of the study target.

We find that because of the ventilation, the estimated NO_x chemical lifetime and emissions decrease as the wind speed increases. The estimated NO_x emission decreases by ~20% from < 3 to 5—7 m s^{-1} wind speed category, and the emission changes little when wind speed is greater than 5 m s^{-1}. Thus the superposition column underestimate NO_x emissions and chemical lifetimes when the wind speed is faster than 5—7 m s^{-1}). The underestimation rate depends on the fraction of days with fast speed, in Wuhan's case, the overall influence is less than 4% for emission and ~8% for chemical lifetimes. We have added this discussion in the revised manuscript in Sect. 3.3.2.

4. Figure 4: Please add error bars to this figure to reflect day-to-day variability. Considering the large variability of emissions and the uncertainties of the model and satellite observations, is the weekly cycle statistically significant?

Response: The reviewer's point is well taken and we have added error bars in the day to day variability of NO_x emissions on each day of the week. We agree with the reviewer that there is no significant weekly cycle on NO_x emissions in Wuhan, and the same finding is also found by the surface NO_2 and O_3 concentration (Wei et al., 2022; Yang et al., 2020) and the traffic flow in Wuhan (https://jtj.wuhan.gov.cn/znjt/zxdt/202409/t20240904_2450210.shtml, last access: 25 November 2024, in Chinese)

5. Section 3.2.3: Considering the large uncertainties of satellite retrievals on daily basis and the potential influences of winds, I think performing the EMG

approach or superposition model over the long-term average data may actually be a better choice for studying the inter-annual variability. I don't see any values added from performing the approach on daily basis. I suggest the authors clarify why it's necessary to calculate daily emissions here.

Response: Thank you for the comment. We agree with the reviewer that there is large uncertainty in satellite retrievals on daily basis, and this is why we did not analyze the variation of NO_x emissions on daily basis, instead we classify the daily emissions into months, seasons, workdays, weekends, and different wind directions and wind speeds. Performing the EMG method over long-term or short term average data is a good choice since there would not be large variation in NO_x emissions during a short time.

However, we argue that it is still necessary to estimate the NO_x emissions on daily basis. First, some unexpected anomaly in NO_x emissions can be identified. For example, Lorente et al. (2019) found highest NO_x emissions on cold weekdays in February 2018 and lowest emissions on warm weekend days in spring 2018, indicating the large contribution from home heating to Paris NO_x emissions. Second, the superposition column model estimate NO_x chemical lifetime and emission through a single overpass of TROPOMI data, avoiding the bias caused by using the averaged NO_2 columns in the nonlinear system (Valin et al., 2013). Third, it is more reasonable to use daily NO_x estimation to infer the co-located CO_2 emission in the city area, because we need the simultaneous and co-located NO_2 and CO_2 observation to ensure an accurate estimation (Zhang et al., 2023).

Please refer to Page 2-3, Line 69-75 in the revised manuscript.

Minor Comments:

Line 48: Please change "ultraviolet/visible" to "ultraviolet (UV)/visible," and use the acronym "UV" for subsequent mentions throughout the manuscript. (Line 85)

Response: Done.

Figure 1b: Better to show the rotated plume with wind direction as x axis, and cross-wind direction as y axis.

Response: Done. Please refer to the Figure 1b in the revised manuscript.

Line 65: EMG model has been used to estimate episodic fire NOx emissions, which does not need long-term average [1]. The key is to find distinguishable plumes from TROPOMI data.

Response: Thank you for the comment, this sentence has been removed.

Line 136: For the title of Figure 1a, please change "origional" to "original." Additionally, could you indicate the location of Wuhan city on the map and include the corresponding radius value?

Response: Done. Please refer to the Figure 1 in the revised manuscript.

References:

[1] Jin X, Zhu Q, Cohen RC. Direct estimates of biomass burning NOx emissions and lifetimes using daily observations from TROPOMI. *Atmos Chem Phys* 2021; 21: 15569–15587.

References:

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Valin, L. C., Russell, A. R., and Cohen, R. C.: Variations of OH radical in an urban plume inferred from NO_2 column measurements, Geophysical Research Letters, 40, 1856-1860, 10.1002/grl.50267, 2013.

Wei, J., Liu, S., Li, Z., Liu, C., Qin, K., Liu, X., Pinker, R. T., Dickerson, R. R., Lin, J., Boersma, K. F., Sun, L., Li, R., Xue, W., Cui, Y., Zhang, C., and Wang, J.: Ground-Level NO_2 Surveillance from Space Across China for High Resolution Using Interpretable Spatiotemporally Weighted Artificial Intelligence, Environmental Science & Technology, 56, 9988-9998, doi: 10.1021/acs.est.2c03834, 2022.

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Zhang, Q., Boersma, K. F., Zhao, B., Eskes, H., Chen, C., Zheng, H., and Zhang, X.: Quantifying daily NO_x and CO_2 emissions from Wuhan using satellite observations from TROPOMI and OCO-2, Atmospheric Chemistry and Physics, 23, 551-563, doi: 10.5194/acp-23-551-2023, 2023.