Reply to reviewer #1

The authors present NO_x emissions in China derived from TROPOMI column measurements and wind data. The dataset matches the scope of ESSD. The paper is generally well written and should be published after dealing with the following issues:

We sincerely thank the reviewer for the constructive and positive feedback on our paper. We have carefully addressed all the comments and revised the manuscript accordingly. Detailed point-by-point responses are provided below, with our replies shown in blue and all revisions in the manuscript highlighted in italics.

Data:

1. Please specify the processor version for the used TROPOMI data. Recent processor updates have made some modifications that results in overall higher tropospheric VCDs, which would affect the discussion of the observed low bias in emissions.

Response: In this study, we use the operational offline TROPOMI NO_2 TVCD product (S5P_L2__NO2___HiR2) from NASA GES DISC. As discussed in Line 494–496, this work primarily aims to propose a practical and insightful perspective for addressing nonlinear NO_x chemistry in emissions estimation, rather than focusing on improvements in retrieval data quality (e.g., air mass factor corrections in satellite NO_2 retrievals). The standard operational NO_2 product is selected, and additional corrections are expected to improve estimation accuracy. We added a brief clarification **in Lines 94–95**: *The operational offline TROPOMI NO_2 TVCD product (S5P_L2__NO2___HiR2) (Van Geffen et al., 2024) from NASA GES DISC (https://daac.gsfc.nasa.gov/datasets/) for 2019–2024 is used in this study.*

Methods:

2. The authors refer to previous work, in particular Ayazpour et al. It is not clear to me how far the current dataset is derived from the method described in Ayazpour, or if modifications/extensions have been made. Please explicitly specify what is new/different in this study as compared to Ayazpour.

Response: The current study builds on the DDA framework by Sun (2022) and its modification by Ayazpour et al. (2025), introducing two main improvements: the application of GEOS-CF chemical data in the modified DDA to better capture strong NO_x gradients near point sources, and the piecewise fitting approach to obtain nonlinear NO_x lifetimes, while the latter represents the main improvement in this study. Figure 3 compares the three DDA-based approaches, illustrating the differences between the original framework, the modified version, and the current study.

We revised the manuscript **in Lines 268–274** as follows: *To illustrate the developments in this study, Figure 3 compares three DDA-based results before and after applying the NO_x/NO_2 ratio correction and improved fitting scheme: (1) a constant NO_x/NO_2 ratio of 1.32 and monthly single-lifetime fitting (fixed_f and single_τ), corresponding to the original DDA framework by Sun (2022); (2) the variable NO_x/NO_2 ratio and monthly single-lifetime fitting (variable_f and single_τ), based on the modification by Ayazpour et al. (2025) and marking the first application of GEOS-CF chemical data in satellite-based emission estimation; (3) a combination of the variable NO_x/NO_2 ratio and piecewise fitting with nonlinear NO_x lifetimes for each month (variable_f and nonlinear_τ), while the latter represents the major improvement in this study.*

3. The lifetime and inverse scale height are fitted based on Eq. (3) for different levels of NO_2 VCDs. However, the basic assumption for this is that emissions are negligible. While this is a good

approximation over remote regions (most parts of WN and WS), I wonder how far this assumption can be made over EN for high NO_2 VCDs. Please extend the discussion accordingly.

Response: As noted by the reviewer , fitting is performed over grid cells with negligible local emissions to isolate the effects of transport, topography, and chemical loss. These locations do not necessarily have low NO_2 columns, as elevated NO_2 TVCDs may result from transport from upwind sources. The fitted scale heights and chemical lifetimes thus reflect effective subregional characteristics rather than strictly local source effects. Piecewise fitting across NO_2 TVCD bins allows pixels with higher NO_2 columns to contribute to the lifetime regression, capturing nonlinear NO_x chemistry; Fig. 1 shows an example of lifetime fitting using high-percentile NO_2 TVCD grids in each subregion. Nevertheless, assuming spatially homogeneous scale heights may lead to overestimation in heavily polluted areas, and lifetimes may still exhibit spatial heterogeneity within each subregion.

We added related discussion **in Lines 480–486**: *The fitted scale heights and chemical lifetimes represent effective subregional parameters derived from grid cells with negligible local emissions, which may lead to overestimation of scale heights in heavily polluted areas and do not fully reflect the sensitivity of NO_x lifetimes to environmental conditions, although the regional averages still capture nonlinear NO_x chemistry. As these are fitting parameters, their physical interpretation should be treated cautiously to avoid over-interpretation (Lonsdale and Sun, 2023). Future observations with higher spatial resolution could improve the representation of scale height and chemical lifetime across heterogeneous regions.*

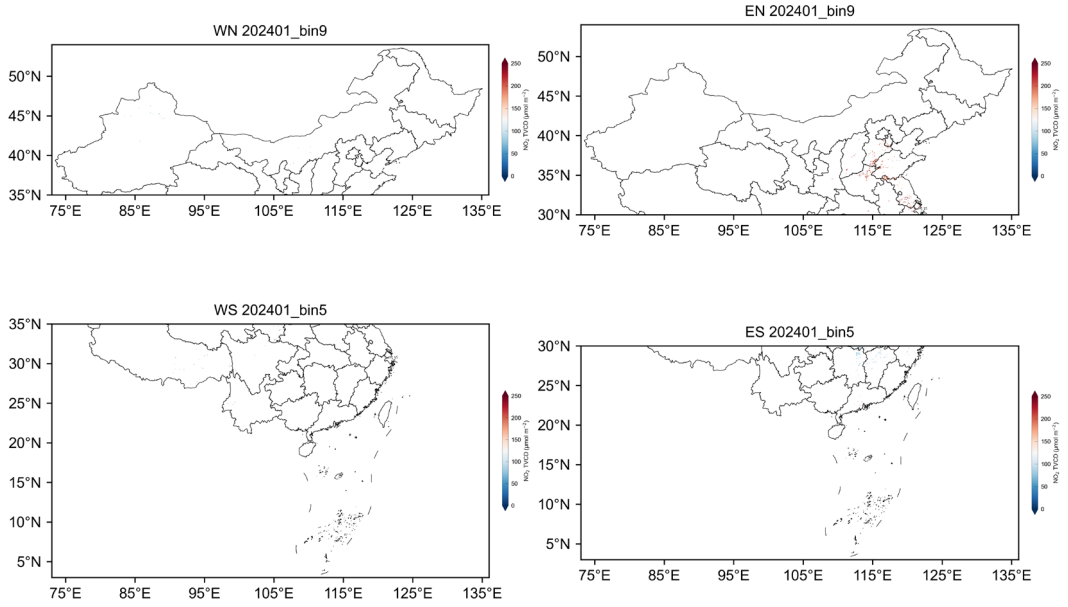

Figure 1. Example of lifetime fitting using high-percentile NO_2 TVCD grids in each subregion, with the number of grids ranging from 48 to 790.

Results:

4. Fitted lifetimes are presented in Fig. 4 and discussed in the text. Please provide and discuss the results for scale height as well. I would expect that scale height increases with distance from source regions, i.e., the assumption of constant X needs to be discussed. I would also expect that X might change seasonally due to different lifetimes.

Response: We added **Fig.S1 to the Supplement** to compare the three DDA-based scale height ($1/X$) for four subregions. The results show clear seasonal variability as well as regional differences. In general,

X is higher in the cleaner and more remote regions (WN and WS) than in the more polluted regions (EN and ES). In the DDA framework, X represents the inverse scale height defined as $X = C|_{z_0}/\Omega$, where Ω is the NO_2 TVCD and $C|_{z_0}$ is NO_2 surface concentration (Sun, 2022). X links the surface concentration to the vertical column density. Since X mainly reflects the effective vertical mixing depth, its variability is primarily controlled by boundary layer mixing rather than chemical lifetime. Consequently, seasonal changes in X are expected and do not necessarily follow the seasonal variation of NO_x lifetime.

We added a brief description **in Lines 324–327**: *In addition, the scale height ($1/X$) fitting results for each subregion are presented in Fig. S1. Scale height shows clear seasonal and regional variability, generally higher in the cleaner and more remote regions (WN and WS) than in the more polluted regions (EN and ES). Its variability is primarily controlled by boundary layer mixing, as X links surface concentration to column density, reflecting the effective vertical mixing depth (Lonsdale and Sun, 2023; Sun, 2022).*

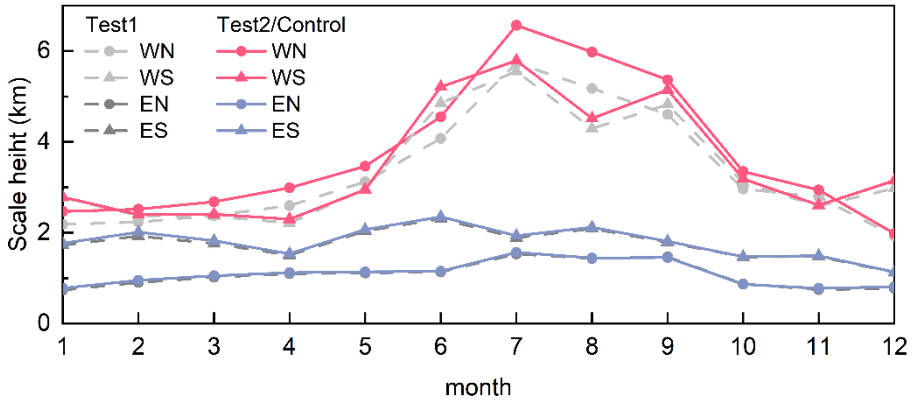

Figure S1. Comparison of monthly scale height (1/X) among three DDA-based setups for each subregion. Test1, Test2, and Control setups are the same as defined in Table S1. The X values are identical for Test2 and the Control setup because both share the same components in the modified DDA framework with variable NO_x/NO_2 ratio.

5. Table S1: Lifetimes for the test setups are far longer that for control, and also far longer than those reported in several recent studies that estimated NO_x lifetimes of the order of 5 hours from (TROP)OMI NO_2 patterns downwind strong sources. What is the reason for the large lifetimes in the test setups? At the same time, fit performance seems to be often better in the test setups than in the control setup. Please comment.

Response: As noted by the reviewer, NO_x lifetimes derived in test1 and test2 are substantially longer than those reported in previous studies. This mainly arises from the fitting strategy. In test1 and test2, a single τ is fitted over relatively clean regions, which tends to overestimate the chemical lifetime. When this single lifetime is applied across the entire study domain, it weakens the effective chemical loss term and introduces an imbalance among local emissions, horizontal transport, and chemical loss. As a result, a considerable number of negative emission grids are reduced, the inferred NO_x flux is underestimated, and the applicability of the lightweight method is limited when extending from point-source to regional emission estimation.

The control setup accounts for nonlinear behavior through piecewise lifetime fitting, yielding shorter and more physically consistent lifetimes. We revised the discussion **in Lines 276–279** to clarify this point as follows: *The nonlinear lifetime fitting more effectively accounts for the balance among local emissions,*

horizontal transport, and chemical loss, reducing negative emission grids and increasing regional emission estimates. Consequently, the improved fitting scheme minimizes artifacts in mountainous and remote regions compared to earlier results (Ayazpour et al., 2025; Beirle et al., 2023; Lonsdale and Sun, 2023; Sun, 2022).

Table S1 compares the lifetime fitting parameters among the three DDA-based setups. In test1 and test2, fitting is applied once over each subregion, yielding a single τ per month. In the control setup, τ is fitted across NO_2 TVCD percentile-based piecewise bins for each month and subsequently averaged for each subregion (Lines 212–226); R^2 and RMSE are the associated fitting statistics referring to the τ fitting. Thus, in Table S1, R^2, RMSE and τ denote the corresponding monthly means across the piecewise bins. For all piecewise bins in EN, R^2 ranges 0.02–0.96, with monthly means in Table S2 of 0.29–0.68; RMSE ranges 0.07–2.3 μmol m^{-2} s^{-1}, with monthly means of 0.25–0.83 μmol m^{-2} s^{-1}; and τ ranges 2.35–17.74 h, with monthly means of 5.10–8.38 h. Compared with test1 and test2, the fit performance of the control setup is more variable across bins, with some bins performing better and others worse. To facilitate comparison, Table S1 lists the monthly means. Overall, the control setup shows more variable performance across bins and higher RMSE than test1/test2, mainly because bins with higher NO_2 TVCDs disproportionately increase the monthly RMSE.

We revised the description of Table S1 **in Lines 5–7 of the Supplement** to clarify this issue: *All values are monthly means: for test1 and test2, the monthly mean is calculated from a single fit over the subregion; for the control setup, the monthly mean is calculated by averaging the piecewise bin results for the subregion. The bottom row shows the mean over the entire study period.*

Dataset:

6. Data is provided on zenodo in form of annual nc files. The data is easily accessible and readable. The following items should be clarified/improved:

Please provide some further information how this data was generated or just add a link to the ESSD paper to the attributes.

Response: We added a link to the ESSD paper on the download page as suggested by the reviewer.

7.Please specify the molecular mass the given emissions are referring to (NO_2?)

Response: As noted by the reviewer, we clarify this issue **in Lines 178–179** as follows and emphasize it in the dataset description: *All bracketed terms are averaged at a monthly scale before X and τ are fitted (see in Sect. 3.2) to derive emissions ⟨EE⟩ in mol m^{-2} s^{-1}, and the conversion to mass assumes NO_x as NO_2.*

8. The unit "tons" is misleading, as this is used differently in e.g., Europe (metric tons) and the US ("short ton"). Please switch to SI units, e.g., 1e3 kg. The given emissions are just given in tons.

Response: The manuscript uses t and kt, referring to metric tonnes (1 t = 1 tonne =10^3 kg; 1 kt = 10^3 tonnes). The dataset annotation uses tonnes, which also denotes metric tonnes. We added a data description as **Table S3 in the Supplement**.

9. From the context, I conclude that these values are referring to "per year" (as there are annual files) and "per pixel". This should be clarified. Due to the link to "pixel", the results depend on the chosen grid, and can NOT be simply interpolated for comparisons to other data. I would thus recommend to switch to emission rate densities (mass per time per area), which then could be easily interpolated and integrated

to any other grid. Then also the unit would be self-explaining. In any case, I recommend to add an additional field "area" (lat coordinate only) which would allow for easy conversion between densities and integrated values by the user.

Response: As noted by the reviewer, the dataset provides gridded anthropogenic NO_x emissions per year per grid (at 0.05° × 0.05° resolution), which represent emission totals rather than emission rate densities. To facilitate potential conversion to emission rate densities (mass per time per area), we added a variable 'grid_area' representing grid-cell area and expanded the dataset description accordingly.

The updated dataset (Version 1.1) is available at https://doi.org/10.5281/zenodo.18923337 (Chen et al., 2026).

We sincerely appreciate the reviewer's thoughtful comments and suggestions, which helped improve both the manuscript and the dataset.

References

Ayazpour, Z., Sun, K., Zhang, R., and Shen, H.: Evaluation of the Directional Derivative Approach for Timely and Accurate Satellite-Based Emission Estimation Using Chemical Transport Model Simulation of Nitrogen Oxides, Journal of Geophysical Research: Atmospheres, 130, e2024JD042817, https://doi.org/10.1029/2024JD042817, 2025.

Beirle, S., Borger, C., Jost, A., and Wagner, T.: Improved catalog of NO_x point source emissions (version 2), Earth Syst. Sci. Data, 15, 3051–3073, https://doi.org/10.5194/essd-15-3051-2023, 2023.

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Sun, K.: Derivation of Emissions From Satellite-Observed Column Amounts and Its Application to TROPOMI NO_2 and CO Observations, Geophysical Research Letters, 49, e2022GL101102, https://doi.org/10.1029/2022GL101102, 2022.