Response to comments by Reviewer #2

We thank Reviewer #2 for the detailed review of our manuscript. The suggestions and comments helped us to further improve the revised version. Below we copy each comment and respond to each of them in blue fonts.

The manuscript "Relation between total-column and near-surface NO2 based on in-situ and PANDORA ground-based remote sensing observations" by Zhang et al. investigates the relationship between total column (TC) and near-surface (NS) NO2 concentrations using field experiments, supported by ancillary data and model analysis. The study highlights the value of Pandora observations in capturing and understanding the dynamic vertical distribution of NO2. Additionally, the use of a backward trajectory model in two case studies effectively illustrates air mass motions at different altitudes, providing meaningful insights into the evolution of TC-NS relationships.

Overall, the study is well-structured and employs a clear methodology. However, concerns regarding its novelty, broader implications, and generalizability may limit its impact. Therefore, major revisions are required to justify its publication in Atmospheric Chemistry and Physics (ACP).

Major comments:

(1) The study is based on a single station (Beijing-RADI) and a short observation period (January 10-29, 2022), which limits the applicability of the results to other regions and seasons. Since the authors emphasize the complexity of the TC/NS NO2 relationship, it raises concerns that this limited dataset may not fully capture its variability. It is suggested that the authors provide a detailed justification for the data selection, explaining why the short-term observations are still appropriate for investigating this relationship. Additionally, the representativeness of the case study should be explicitly discussed, particularly whether the findings are expected to hold across different meteorological conditions and locations. If possible, a comparison of the TC-NS NO2 relationship between the Beijing-RADI site and other sites is recommended to strengthen the generalizability of the conclusions.

Response: Thanks for your comment. We agree with your point, yet our considerations are as follows: firstly, Beijing was selected as the study area due to the persistent pollution in the Beijing-Tianjin-Hebei (BTH) region, where polluted days accounted for 36% of the year in 2020, with 3% of these days experiencing heavy pollution (China Ministry of Ecological Environment, 2020). This highlights the importance of research in this area. Secondly, Beijing, as the capital of China, is a megacity with a vast population. Surrounded by mountains to the northwest, polluted weather forms easy during southwest, southeast, or calm wind conditions. Consequently, many studies have focused on Beijing or the urban agglomeration of the BTH region. In recent years, based on research on fine particulate matter, the types of pollutant transport in the Beijing area have been deeply investigated. For instance, Yin et al. (2025) indicated that there are three pollution types for Beijing: SW, SE, and SM. The SW type involves pollutants being transported from Shanxi through Hebei to Beijing, while the SE type primarily involves pollutants being transported from Shandong and the eastern side of Hebei, as well as Tianjin. Other studies (Dong et al., 2020; Chang et al., 2019; Li et al., 2017) have also reached consistent conclusions. Most of their analyses are based on multi-year pollution classification in the Beijing area and are representative, laying a foundation for our research. Given limited experimental funding and conditions, we selected January, the winter season in Beijing, as the study period based on the patterns summarized in these studies. We conducted comprehensive ground-based measurements using various instruments, complemented by the first Pandora spectrometer in China and satellite observations, developed the study of vertical pollution and regional transport characteristics in Beijing. As previous studies have reported the contributions of different pollution transport types to the Beijing area, our objective is to identify representative cases to illustrate the impact of different transport types on the vertical structure of NO_2 and its diurnal evolution patterns. This can provide scientific explanations for the increasing number of satellite remote sensing studies on NS NO_2 concentrations and also suggest that future research using geostationary satellites to study NS NO_2 concentrations should focus on vertical variations within the daytime boundary layer. We appreciate the comment of expanding observations (time period and stations), but due to observational limitation (Pandora and Lidar are expensive instruments), this is currently not feasible. In the future, we will strive to develop as many comprehensive observation stations as possible and collect more representative cases to further address the scientific questions focused on in this study. In order to further explain our ideas, we have made the following modifications to the MS:

Line 403-410: “Many studies (Yin et al., 2025; Dong et al., 2020; Chang et al., 2019; Li et al., 2017) indicated that the pollution in Beijing mainly originates from three pollution transport patterns: southwest (SW), southeast (SE), and south mixing (SM) patterns. Among them, the most representative are the SW transport path from Shanxi province to Shijiazhuang-Baoding-Beijing and the SE transport path from Shandong province to Cangzhou-Langfang-Tianjin-Beijing. However, few studies have explored the vertical transport of pollution types. Cases 1 and 2 are representative for the widespread SW and SE patterns, respectively.”

(2) The study emphasizes the importance of investigating the TC-NS NO2 relationship for satellite-based monitoring of NS NO2 concentration and discusses how the TC/NS ratio changes with time and meteorology. However, the manuscript does not directly analyze the relationship between satellite TC NO2 and NS NO2, instead focusing solely on the Pandora TC NO2 and NS NO2. Given that the authors acknowledge biases between Pandora and satellite TC NO2 in the introduction, it is recommended that additional analyses be included to assess how TC/NS variations impact actual satellite NO2 applications on NO2 monitoring.

Response: Thanks for your comment. First, to avoid ambiguity, we refer to TC NO_2 as the total vertical column density (VCD) of NO_2, and the manuscript has been thoroughly revised accordingly. For polar-orbiting satellite observations, we have compared the total VCD and tropospheric NO_2 concentrations obtained by TROPOMI and Pandora from August 2021 to July 2022 in our previous study (Liu et al., 2024) and found a good correlation (Fig. R1), as mentioned in the Introduction (Lines 96-99). Figure R1 demonstrates that the correlation coefficient R between TROPOMI and PANDORA for both the total VCD of NO_2 and for the tropospheric VCD of NO_2 exceeds 0.9. In comparison, the mean difference (MD) between total VCD of NO_2 concentrations from TROPOMI and Pandora is 8.44%, while tropospheric NO_2 concentrations exhibit an MD of 16.15%. For absolute differences, the total VCD has a slightly higher value than the tropospheric results, with mean absolute differences (MAD) of 3.14 and 2.71 Pmolec cm^{-2}, respectively. For error distribution (Fig. R1b&d), the tropospheric NO_2 is closer to a normal distribution, while the mean error of total VCD of NO_2 shows a shift of -1.16 Pmolec cm^{-2}.

It is noteworthy that during the field experiment period in January 2022, we obtained observations from a Trace Gas Analyzer and a Lidar, but there were no total VCD of NO_2 products available from neither TROPOMI nor GEMS during that same period. This posed a certain obstacle to our research. However, the main objective of this study is to capture the vertical distribution changes of golden cases and understand the vertical distribution differences. Therefore, we believe that the comparison of satellite data can be deferred to the next stage of research, and this stage should focus on analyzing the temporal-spatial complexity of NO_2. Fortunately, we did identify two such cases during the field experiment. Although these cases do not cover all the major pollution transport types (SW, SE, SM) mentioned in previous studies (e.g. Yin et al., 2025) in Beijing, we have explained the temporal evolution of horizontal and vertical pollution within the limited observations. We believe that this is meaningful.

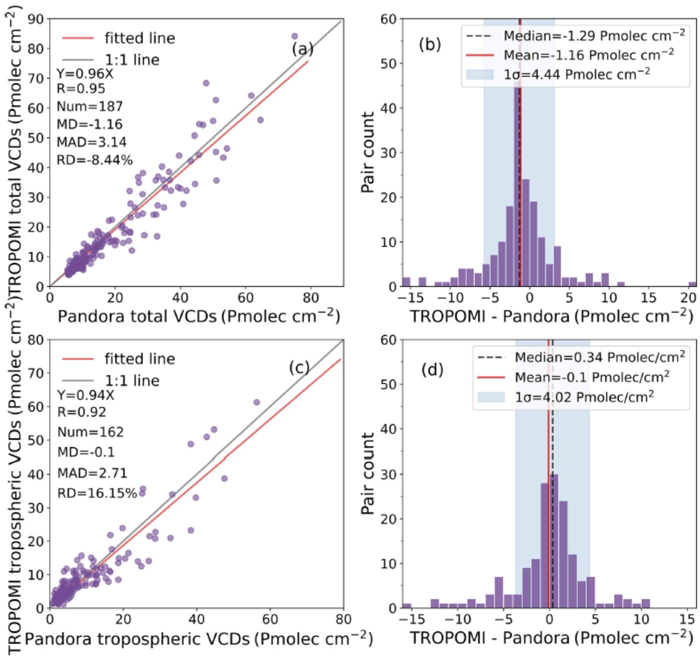

Figure R1. Validation of TROPOMI total and tropospheric NO2 VCDs, resampled to a spatial resolution of 100m×100 m, using Pandora observations as reference data: (a, c) scatterplots of total and tropospheric TROPOMI vs. Pandora data together with statistical metrics; (b, d) histograms of the differences between TROPOMI and Pandora NO2 total / tropospheric VCDs. (Liu et al., 2024, Figure 9)

(3) The manuscript does not clearly establish the novelty of the study compared to prior works. The motivation expressed in lines 145-148 is not sufficiently developed to justify the study's significance. While the paper states that accurate TC/NS NO2 information was not previously available for China, it does not explicitly explain why this makes the study novel or how it differs from previous research on TC/NS relationships. To strengthen the motivation, the authors should clarify what specific gaps in the literature they are addressing and explicitly compare their approach to existing studies.

Response: Thanks for your comment. I am very sorry that we didn't clearly articulate the innovation of the research. We believe that the rapid advancement of satellite remote sensing technology currently enables regional detection of atmospheric pollutants. However, satellite remote sensing primarily captures the pollutant content within the atmospheric column, whereas it is the near-surface pollution that directly impacts human health. Numerous studies (Chang et al., 2025; Wei et al., 2022; Zhang et al., 2022; Dou et al., 2021) have been conducted on converting the pollutant content in the atmospheric column obtained by satellite remote sensing into near-surface concentrations. Yet, a crucial issue arises: the complex evolution of the local boundary layer limits the accuracy of estimating near-surface pollutant concentrations. Therefore, our research aims to utilize ground-based remote sensing observations and near-surface NO_2 concentration measurements to investigate vertical decoupling of NO_2 within the boundary layer during winter in Beijing and discuss the influence of regional pollutant transport on the vertical distribution of NO_2 concentrations. Although there are numerous studies on the vertical distribution of NO_2 (Sun et al., 2023; Zhang et al., 2023; Kang et al., 2021) and the contribution of regional transport (Yin et al., 2025; Dong et al., 2020; Chang et al., 2019; Li et al., 2017), respectively, research linking regional pollutant transport to the vertical distribution of NO_2 concentrations and clarifying the impact of regional transport on vertical concentration distributions remains scarce. Focusing on this key aspect, we propose possible connections between regional pollutant transport and vertically stratified NO_2 concentrations, with the aim of providing valuable information for satellite remote sensing to predict near-surface NO_2 concentrations. We have added the following text to Section 1, describing gaps that are addressed in this study and resulting innovations and value of this study:

Line 146-166: "While geostationary satellites enable continuous daytime observations of total VCD of NO_2, discrepancies between total VCD and NS concentrations of NO_2 concentrations remain a critical challenge. The weak correlation between NS NO_2 concentrations and satellite-derived total VCD of NO_2 (Lamsal et al., 2014) is closely tied to differences in their vertical distribution, atmospheric lifetimes, and chemical reaction pathways (Xing et al., 2017). Despite extensive efforts to derive NS NO_2 concentrations from total VCD measurements (Chang et al., 2025; Wei et al., 2022; Zhang et al., 2022; Dou et al., 2021), the dynamic complexity of the planetary boundary layer introduces substantial uncertainties. Moreover, prior studies have emphasized the roles of vertical NO_2 distribution (Sun et al., 2023; Zhang et al., 2023; Kang et al., 2021) and regional pollutant transport contributions (Yin et al., 2025; Dong et al., 2020; Chang et al., 2019; Li et al., 2017), but research explicitly linking regional transport processes to vertical NO_2 concentration gradients and elucidating their interactive effects remains limited. Song et al. (2024) obtained NS NO_2 concentrations based on the Himawari-8 geostationary satellite using machine learning, which has good performance in the noon and afternoon, and relatively poor performance in the morning. These knowledge gaps are further exacerbated by satellite data limitations in resolving NS pollution, which has direct implications for human health assessments. To address these challenges, we need to integrate ground-based remote sensing observations with in situ NS NO_2 measurements to investigate vertical decoupling phenomena, and investigate the influence of distinct pollutant transport pathways on NS NO_2 pollution dynamics."

(4) The manuscript presents an analysis of the TC/NS NO2 ratio, but its physical significance and relevance to satellite-based NO2 applications remain unclear. Besides, the sensitivity of the ratio to meteorological factors is not quantified. To strengthen the analysis, the authors should clarify the rationale for using this ratio, ensure a consistent interpretation, assess meteorological influences, and explicitly connect their findings to the use of satellite NO2 data for estimating near-surface NO2

Response: Thanks for your comment. We apologize for any confusion caused by our unclear statement. From the analysis of the two cases, we observe a high correlation coefficient R, suggesting that the relationship between total VCD and NS NO_2 concentrations can be characterized by y = ax + b. However, in a larger number of cases, the correlation is less ideal, particularly during the morning hours. The low correlation does not quantify the changes between total VCD and NS, prompting us to introduce the Ratio as a means of quantification. Furthermore, the Ratio can also express the degree of dispersion between total VCD and NS to a certain extent; that is, the more unstable the Ratio, the higher the dispersion and the poorer the correlation. In contrast, the Ratio is generally more stable after 13:00, indicating the feasibility of using polar-orbiting satellites to predict NS NO_2 based on total VCD of NO_2 after this time. However, using geostationary satellites for the same prediction based on total VCD observations before 13:00 poses greater challenges. Notably, the analysis of case 1 and case 2 has already shown that regional pollution transport paths may be helpful for predicting NS NO_2 in winter in Beijing. In other words, considering both the spatial distribution of pollutants and their transport direction in predictions has the potential to enhance the ability of satellites to predict NS NO_2 concentrations based on total VCD of NO_2. The limitation of our study is that it is based on only one month of observations, and during this period, the geostationary satellite GEO-KOMPSAT-2B /GEMS did not release any products, which prevents us from better demonstrating the universality and applicability of our findings. In future research, we will obtain more observations and utilize geostationary satellite observation products to further refine and validate our discoveries.

We further elaborated on the role of Ratio in the MS as follows:

Line 706-712: "The Ratio, defined as the ratio of total to NS NO_2 concentrations, serves a dual purpose: it not only quantifies the changes between total VCD and NS concentrations of NO_2 when the correlation is low but also reflects the degree of dispersion between the two measurements. A more variable Ratio indicates higher dispersion and poorer correlation, providing a straightforward yet effective way to assess the reliability of using total VCD of NO_2 to predict NS NO_2 concentrations."

We further summarized the comprehensive point presented by the analysis of Ratio and the two cases as follows:

Line 737-749: "Generally, the temporal stability of the Ratio is important. The Ratio is overall less variable after 13:00, suggesting that polar-orbiting satellites can be used to predict NS NO_2 based on total VCD of NO_2 during this period with greater confidence. This temporal stability is particularly valuable because it offers a feasible approach for air quality monitoring and forecasting. In contrast, the Ratio is less stable before 13:00, posing greater challenges for using geostationary satellites for the same prediction task. It's worth noting that our analysis in winter in Beijing suggests that considering both the spatial distribution of pollutants and their transport direction has the potential to enhance the ability of satellites to predict NS NO_2 concentrations based on total VCD of NO_2. By incorporating this information into prediction models, the accuracy and reliability of satellite-based air quality predictions may be improved, particularly in complex urban environments where pollutant concentrations can vary significantly over short distances and time periods."

Further, the reviewer asks for "sensitivity of the ratio to meteorological factors is not quantified" and "assess meteorological influences". Obviously, a case study for a limited period of time, including a period when the meteorological situation did not allow for collecting the full set of data (period 2, due to the occurrence of clouds), is not suited for such analysis which requires a large variety of meteorological situations. Rather, we have identified the problem based on the analysis of a comprehensive set of data including ground-based in situ and remote sensing, satellite remote sensing, air mass trajectory analysis and meteorological observations, which are usually not available form the same site. Furthermore, as explained in the response to comment 6 for reviewer #1, satellite data were not available during the time of the field study and thus "explicitly connect their findings to the use of satellite NO_2 data for estimating near-surface NO_2" is not possible. We regret that we are currently unable to utilize meteorological factors for quantitative assessment of Ratio in this study. In our future research, we will focus on this aspect, with the aim of establishing a connection between Ratio and satellite-observed total VCD of NO_2 concentrations, and making practical contributions to near-surface NO_2 predictions.

Specific comments:

(1) Lines 58-60: The phrase 'providing vertical total column and tropospheric densities' is unclear. It would be more precise to explicitly distinguish total column density, stratospheric column density, total and tropospheric column density. In this manuscript, 'TC' should consistently and explicitly refer to the 'tropospheric vertical column density' to avoid ambiguity.

Response: Thanks for your comment. We modified this sentence as follows:

Line 57-59: "Concentrations of NO_2 in the atmosphere can be measured using satellite-based sensors providing total and tropospheric column densities, ground-based remote sensing using MAX-DOAS or Pandora instruments, or in situ instruments."

Additionally, in this study, total VCDs refers to total column, which is derived from the nvs3 product of Pandora, while partial column NO_2 is obtained from the uvh3 product. We clarify this in the original text as follows:

Line 224-225: "In view of this high precision, we use total VCD of NO_2 from the nvs3 product in this study and select data with quality control flag of L10."

Line 237-239: "The NO_2 of the partial column can be obtained from the uvh3 product which was downloaded from the PGN website (https://pandonia-global-network.org, last accessed: 22 Jan 2025)."

(2) Lines 85-88: Thompson's work focuses on the complexity of the TC-NS NO2 relationship, which does not align well with the paragraph's main discussion on Pandora vs. satellite TC NO2 This reference would be more appropriate in a section specifically addressing TC-NS variability rather than in a discussion of measurement consistency between Pandora and satellite data.

Response: Thanks for your comments. We have moved Thompson's work to the next paragraph to discuss the complex relationship between total VCD and NS NO_2.

Line 113-119: "… Similarly, Liu et al. (2024) show different relations between total VCD and NS concentrations of NO_2 for low and high concentrations which are qualitatively explained in terms of transport and local emissions. Moreover, Thompson et al. (2019), using data from the KORUS-AQ coastal cruise experiment, reported that there is no consistent correlation between total VCD and NS concentrations of NO_2 across different cases and that the relation between total VCD and NS concentrations of NO_2 is complex."

(3) Lines 109-114: This sentence is excessively long, making it difficult to follow. It is recommended to split it into two sentences to improve readability and ensure clarity of the message.

Response: Thanks for your comment. We have modified this sentence as follows:

Line 107-113: "Their results indicate that total VCD and NS concentrations of NO_2 exhibit a stronger correlation under advective boundary layer conditions at high wind speeds, where the vertical distribution of NO_2 is more uniform. In contrast, in the presence of plumes from large point sources, either decoupled from the surface or transported from nearby cities, enhance the vertical heterogeneity of NO_2. These plumes contribute to a less consistent relationship between total VCD and NS concentrations of NO_2."

(4) Figure 2: It is suggested to add dashed lines in Figure 2 to clearly indicate the boundaries between the three periods, improving readability.

Response: Thanks for your comment. We have added the boundaries between the three periods using dash lines in the Figure 2.

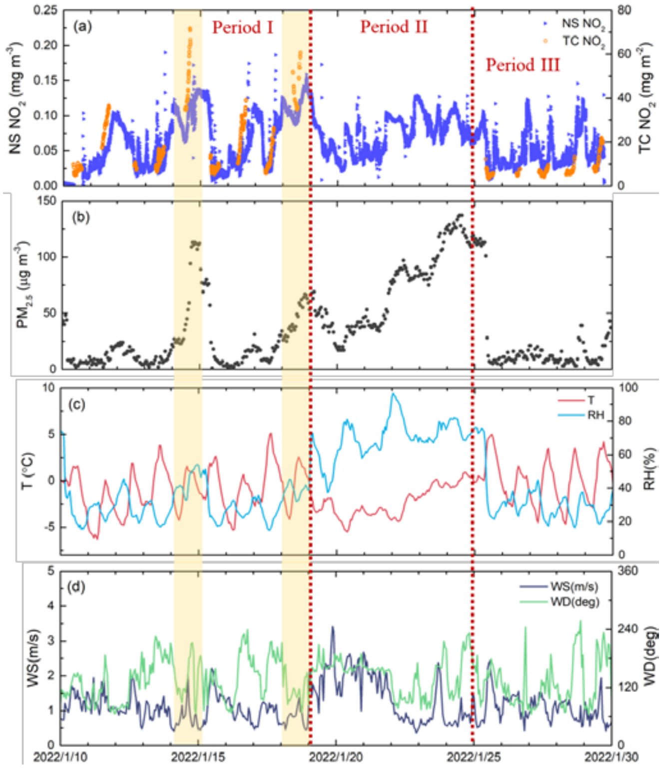

Figure 2. Time series of observed parameters from Jan 10 to 29, 2022 (a) total VCD and NS concentrations of NO_2 concentrations, (b) NS $PM_{2.5}$ concentration, (c) temperature and related humidity, and (d) wind speed and wind direction from WMO meteorological station 54511 in Beijing. The vertical dotted lines mark the boundaries between the three periods and the yellow shaded rectangles mark the two cases discussed in Section 3.2.

(5) Lines 315-322: The manuscript presents PM2.5 observations and results but does not explicitly explain their relevance to NO2 analysis. To improve clarity, the authors should justify the inclusion of PM2.5 data and clarify how it supports the study's objectives, ensuring a stronger connection between NO2 assessment and aerosol pollution.

Response: Thanks for your comment. The $PM_{2.5}$ mass concentration observations serves two purposes: (1) NS pollution in the Beijing area is usually not dominated by NO_2, but rather by $PM_{2.5}$. Also, the air mass trajectories for case 2 the increased wind speed at 16:00, which marks the end of the pollution episodes. In fig 2 we see that clearly in both the NO_2 and $PM_{2.5}$ concentrations, so we use $PM_{2.5}$ as an additional indicator. (2) Since our vertical information comes from Lidar, and the Lidar signal is caused by the backscattering from particulate matter, the NS $PM_{2.5}$ mass concentration serves as auxiliary information for Lidar observations. Therefore, we believe that the $PM_{2.5}$ observations is necessary. We have also utilized $PM_{2.5}$ information on multiple occasions, for example:

Line 495-497: "The increase of the NS concentrations is consistent with the highest $PM_{2.5}$ concentrations as presented in Fig. 2b and the overall increase of the lidar signal, indicating increasing aerosol concentrations."

Line 606-608: "The lidar data in Fig. 5c, with lower intensity than on 14 January, indicate smaller aerosol concentrations on 18 January than on 14 January, consistent with the smaller $PM_{2.5}$ concentrations (in Fig. 2b)."

(6) Line 484: The TC/NS ratio mentioned in Line 484 is not found in Figure 3b. Please clarify whether it is missing or referenced incorrectly.

Response: Thanks for your comment. We apologize for this mistake. Instead of "ratio," it should be "relationship." We have revised this sentence as follows:

Line 525-528: "As a result, the temporal variation of the concentrations in both layers was in part influenced by the same processes, differences were not large (Fig. 3a) and the relationship between the total VCD and NS concentrations was linear with a small slope (174.24) and well-correlated (R=0.94) (Fig. 3b)."

(7) Figure 3: There appear to be unexpected content between Figures 3c and 3d, likely due to figure cropping. Please check and correct this issue.

Response: Thanks for your comment. We have updated the figure 3 and 5 as follows:

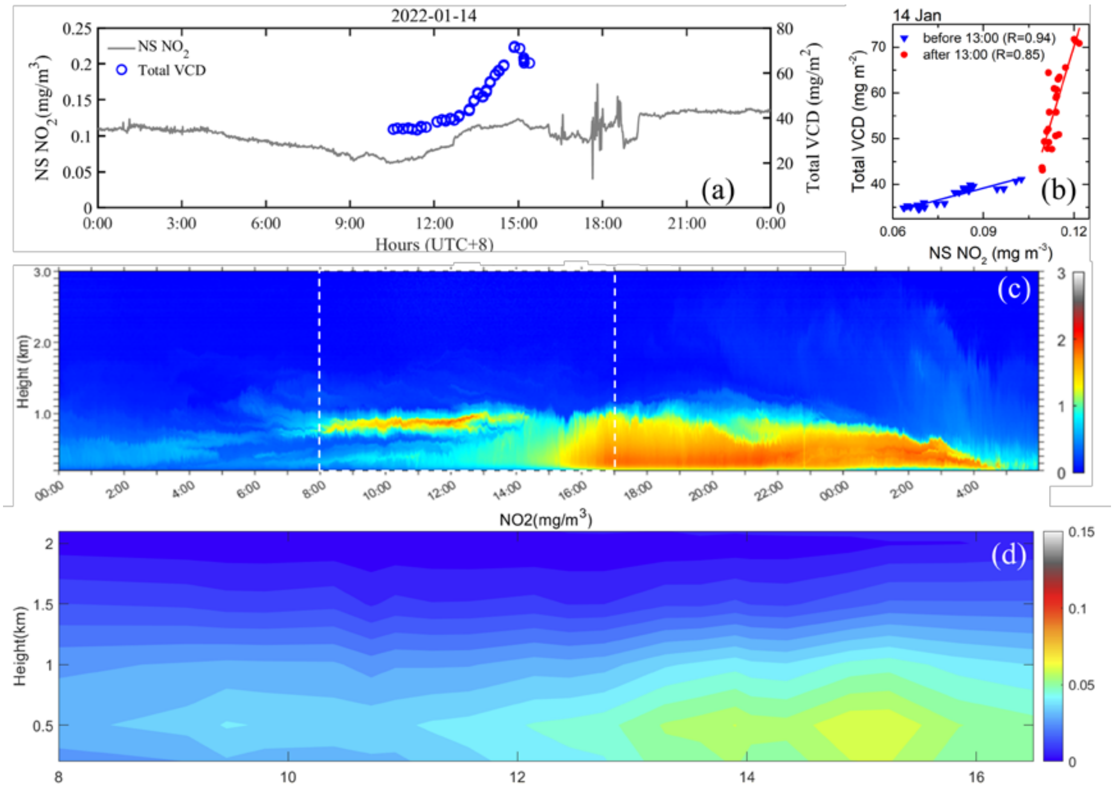

Figure 3. (a) Time series of NS NO_2 (grey line) and total VCD of NO_2 (blue circles) at the Beijing

RADI site (40.004°N, 116.379°E) on Jan 14, 2022; (b) scatterplots of total VCD and NS concentrations of NO_2 and fits to these data during the morning (before 13:00) and during the afternoon (after 13:00), showing different relationships as discussed in the text; (c) time series of vertical profiles of range-corrected lidar signal at 1064 nm. Note that the lowest height in Fig. 3c is 100 m; (d) time series of NO_2 vertical profiles derived from Pandora sky radiance measurements. Note that the Pandora profiles are constructed from layer-averaged volume mixing ratios interpolated to 6 standard levels and the lowest level is 0.2 km.

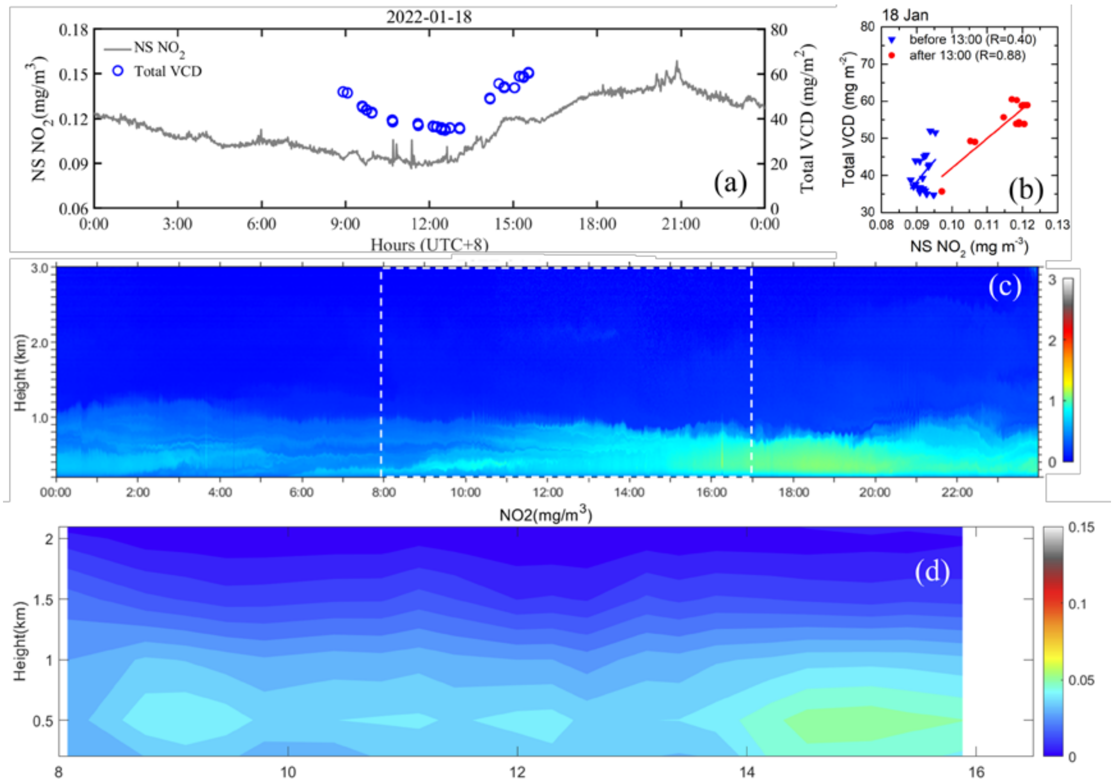

Figure 5. Same as Fig. 3 but for 18 January, 2022.

(8) Figure 4: To enhance clarity, it is suggested to add necessary subtitles for the three subfigures in Figure 4c, such as local time. Additionally, the resolution of Figure 4 appears low and should be improved. (Same for Figure 6)

Response: Thanks for your comment. We have added the subtitles for the subfigures and improved the resolution of Figure 4 as follows:

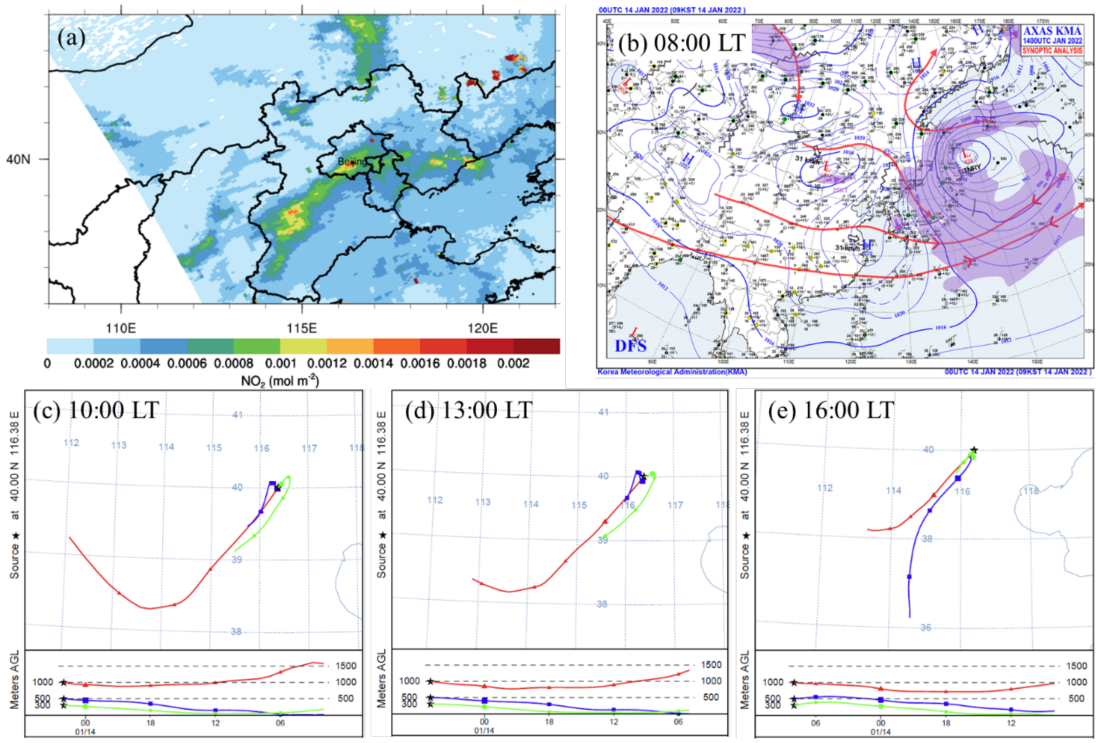

Figure 4. (a) Spatial distribution of tropospheric NO_2 in the study area derived from TROPOMI data on 14 January 2022; (b) Synoptic weather map at 00:00 UTC (08:00 LT); 24-hour backward air mass trajectories arriving at the Beijing-RADI site at (c) 10:00, (d) 13:00 and (e) 16:00 LT, at heights of 300, 100 and 1000 m, calculated using the HYSPLIT model with 6h time steps (00, 06, 12 and 18) and a shorter time step to the arrival time.

(9) Figure 6: The middle panel of Figure 6c contains unexpected content. Please verify and correct this issue.

Response: Thanks for your comment. We have added the subtitles for the subfigures and updated the Figure 6 as follows:

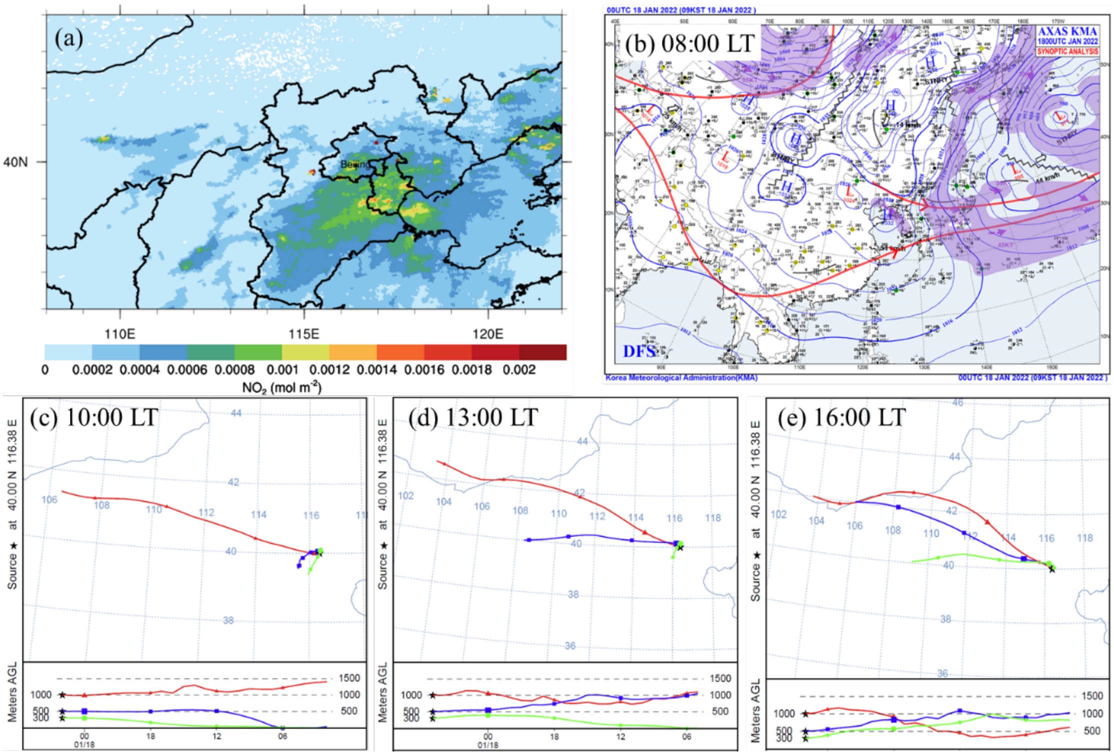

Figure 6. Same as Fig. 4, but for 18 January, 2022.

(10) Lines 691-693: The outlook on potential large-scale implementation is vague. The authors should specify which shortcomings to be resolved.

Response: Thanks for your comment. We further pointed out the limitations of the study and revised the manuscript as follows:

Line 796-799: "Moreover, we will broaden the scope of experimental areas and field sites to complement research on the various pollutant emission and transport characteristics. Furthermore, observations over a longer period will allow us to capture more representative cases, thereby enhancing the reliability of our findings."