Thank you for your careful review and constructive suggestions. These suggestions are quite valuable to us, and help improve our manuscript a lot.

Point-to-point responses

We appreciate the reviewers for their valuable and constructive comments, which are very helpful for the improvement of the manuscript. We have revised the manuscript carefully according to the reviewers' comments. We have addressed the reviewers' comments on a point-to-point basis as below for consideration, where the reviewers' comments are cited in ***black****, and the responses are in* ***blue****.*

General Comments:

The manuscript entitled "Evaluation of Transport Processes over North China Plain and Yangtze River Delta using MAX-DOAS Observations" by Song et al investigates the transport patterns and vertical distributions of NO2, HCHO and aerosols using a number of instruments from a MAX-DOAS network in China. The temporal variation of the air mass composition has been investigated using modelled wind fields, which allow to identify air masses moving from the region of one instrument to the other. This is a very useful approach that allows to investigate the dynamical and chemical processing of individual air masses.

A main problem of the manuscript is that there are many occasions where no clear distinction has been made between (1) conclusions evidently inferred from the measurements, (2) findings from other studies that support the measurements and (3) hypotheses based on the observations. Many conclusions drawn from measurements are highly speculative, for example that the presence of NO2 and HCHO enhance the AEC, and that secondary aerosols are present (Section 3.1). Another example is the statement "We discovered that secondary aerosol generation always accompanied the regional transport process" (L302), for which there is neither direct evidence from the measurements nor any other study mentioned that would support this finding.

Re: Thank you for this comment. There are many speculative arguments making Section 3.1 confusing and less convincing. In addition, our descriptions jump between Fig. 3 and Fig. 4, which easily cause misunderstanding. To make Section 3.1 more understandable and emphasize the main points, we have deleted many speculative conclusions (e.g., secondary aerosol generation, aerosol transport among stations) and changed the structure of Section 3.1 as follows.

"Figure 3 presents the temporal variations in the vertical distributions of aerosols, NO_2, and HCHO during this regional transport. At the CAMS and NC stations, the aerosol, NO_2, and HCHO concentrations were consistently high near the surface, primarily because of the heavy traffic flow and dense factory emissions in Beijing (Zhang et al., 2016; Li et al., 2017). Previous studies have suggested that urban air pollution in Beijing is dominated by a combination of coal burning and vehicle emissions, which results in severe particulate pollution (Wang and Hao, 2012; Wu et al., 2011). At SJZ, the NO_2

concentration was high (~ 12 ppb) in the morning and late afternoon, whereas the concentration was lowest (~ 6 ppb) near noon, which is explained by the morning and evening rush hour. Comparatively, the overall AEC and NO_2 levels were relatively low at the WD station, whereas a continuous high-value HCHO distribution (> 2 ppb) occurred at 0-1500 m between 11:00 and 16:00. This occurred because the WD station is located in a farm field with less traffic flow and high vegetation coverage; therefore, large amounts of HCHO are directly emitted by biogenic sources and secondarily produced by natural and anthropogenic volatile organic compound (VOC) photolysis (Wang et al., 2016; Wu et al., 2017a).

Nevertheless, Fig. 3 cannot reveal the exact layers in which the main transport phenomena occur. For instance, at the CAMS station, the AEC at the surface and upper layers both reached ~ 0.5 km^{-1} around noon, making it difficult to determine the layer in which transport was more obvious. To further demonstrate the dynamic transport process of different pollutants, we calculated the hourly F_i and F_c, and defined the MTL. As shown in Fig. 4, a positive F_i indicates that all transport flux projections in the southwest-northeast pathway are from southwest to northeast at the four stations. The MTLs of aerosols, HCHO, and NO_2 exhibited different spatiotemporal characteristics. Although surface and high-altitude (400–800 m) AECs both remained at a relatively high level (> 0.3 km^{-1}) at CAMS during 12:00-17:00 (Fig. 3), there was a large discrepancy between their corresponding F_i values (Fig. 4). The aerosol near-surface F_i was ~ 1 $km^{-1}\cdot m\cdot s^{-1}$ after 12:00, while F_i in layers of 400–800 m all exceeded 1.2 $km^{-1}\cdot m\cdot s^{-1}$, and even reached ~ 2 $km^{-1}\cdot m\cdot s^{-1}$ around 12:00. At the SJZ station, the AECs at surface and 300–1000 m layer mostly ranged from 0.3 to 0.4 km^{-1}, especially after 10:00 (Fig. 3). However, the MTLs of aerosols were mostly at 400–800 m throughout the day, with many transport fluxes in those layers even reaching ~ 2 $km^{-1}\cdot m\cdot s^{-1}$ (Fig. 4). At the WD station, the highest F_i also tended to occur at high layers (400–800 m), with maximum F_i exceeding 1.7 $km^{-1}\cdot m\cdot s^{-1}$ at 400–500 m at 15:00. This suggested that aerosol transport occurred mainly in the upper layers. In the late afternoon, aerosols gradually accumulated towards the surface, and triggered a variation in the distribution of F_i. After 16:00, the shift in the high-AEC air mass caused the transport fluxes in the lower layers (100–200 m) to increase to > 1.1 and ~ 2 $km^{-1}\cdot m\cdot s^{-1}$ for the CAMS and SJZ stations, respectively. Surface aerosol accumulation is closely linked to the collapse of the mixing layer and formation of a stable nocturnal boundary layer (Ding et al., 2008; Ran et al., 2016). Remarkably, high-altitude aerosol air masses began to mix with near-surface aerosols after 14:00 at the NC station (Fig. 3), triggering a variation in the MTL (Fig. 4). This might be explained by enhanced vertical mixing due to the heating of the surface during the course of the day (Castellanos et al., 2011; Wang et al., 2019). Generally, the MTL of aerosols was situated at 400–800 m during the daytime, where variations in the boundary layer and increased vertical mixing can influence the MTL. In contrast to aerosols, we found that a high-value NO_2 F_i frequently occurred in the 0–400 m layer. Except that F_i reached the highest level of ~ 50 $ppb\cdot m\cdot s^{-1}$ in the 400–600 m layer at 16:00 at the SJZ station, the other highest F_i all occurred below 400 m at any station and at any time. This indicated that the MTL of NO_2 was 0–400 m. Near-surface NO_2 emission sources (e.g.,

vehicle and factory emissions) might be the main reason for this phenomenon. Compared with aerosols and NO_2, we found that high-value HCHO F_i extended to higher altitudes. Taking CAMS as an example, we found the strongest HCHO F_i constantly emerging at 1000–1200 m from 8:00 to 13:00, and averaging 9.18 ppb·m·s^{-1}. During the same period, surface HCHO F_i only averaged 6.44 ppb·m·s^{-1}. However, at the CAMS station, the surface HCHO concentration was much higher than that of the 1000–1200 m layer between 8:00 and 13:00 (Fig. 3), proving that high-altitude transport contributed more to overall HCHO transport. After 10:00, we found that the highest HCHO F_i gradually increased from ~ 8 to ~ 20 ppb·m·s^{-1} at WD, with the MTL of HCHO ranging from 400 to 1000 m. At station SJZ, the strongest HCHO F_i increased from ~ 10 to ~ 16 ppb·m·s^{-1} during 11:00–17:00, with the highest transport fluxes occurring mostly at 400–800 m. These findings indicated that the MTL of HCHO was mainly 400–1200 m. The sharp variation in the MTL at the NC station might be caused by atmospheric vertical mixing (Castellanos et al., 2011; Wang et al., 2019). As shown in Fig. 3, high HCHO concentrations tend to appear at higher altitudes than those of aerosols and NO_2. A possible explanation is that the precursor compounds of HCHO are transported to higher layers and converted into HCHO through photochemical reactions, resulting in elevated HCHO concentrations at higher altitudes (Kumar et al., 2020). Furthermore, strong high-altitude winds were more conducive to HCHO transport (Fig. S5), which further increased the corresponding transport flux. Notably, HCHO F_i was enhanced around noon because the increased solar radiation promotes the secondary generation of HCHO. Long-term observations have revealed that secondary HCHO formation through VOCs photolysis plays a significant role in Beijing (Liu et al., 2020; Zhu et al., 2018).

In addition, we discovered a wide discrepancy in the F_c among stations for various pollutants (Fig. 5). The average aerosol F_c decreased in the following order: SJZ (3.21×10^3 km^{-1}·m^2·s^{-1}) > NC (2.69×10^3 km^{-1}·m^2·s^{-1}) > CAMS (2.43×10^3 km^{-1}·m^2·s^{-1}) > WD (1.42×10^3 km^{-1}·m^2·s^{-1}). For NO_2 transport, the average F_c values at SJZ (5.69×10^4 ppb·m^2·s^{-1}), NC (4.42×10^4 ppb·m^2·s^{-1}), and CAMS (6.16×10^4 ppb·m^2·s^{-1}) were substantially higher than those at WD (2.04×10^4 ppb·m^2·s^{-1}). Conversely, the average F_c of HCHO was the highest in WD (3.21×10^4 ppb·m^2·s^{-1}), whereas the F_c values in SJZ, NC, and CAMS were 1.76×10^4, 2.01×10^4, and 1.94×10^4 ppb·m^2·s^{-1}, respectively. In terms of the relative locations of stations (Fig. 1) and the F_c results, we considered that SJZ was an important source of transported aerosol and NO_2, and WD was one of the main HCHO sources during this regional transport, which largely affected the air quality of cities along the southwest-northeast transport pathway. The corresponding error distributions of F_i and F_c were provided in Figs. S6 and S7."

I suggest to remove the discussion on dust properties inferred from the measured intensity. MAX-DOAS instruments are usually not radiometrically calibrated. Even if the spectrometers are of the same type, the signals from different instruments cannot be directly compared to each other since they depend on many parameters, such as the gain of the amplifier, as well as on the adjustment of the telescope optics, the length of the fibre bundle, etc. I therefore suggest to remove the corresponding paragraphs (Fig. 6 and L368-374), which anyway do not provide much extra information compared to the

retrieved extinction profiles.

Re: Thank you for this comment. We have given up classifying the four stations and basing the following discussions on the classification here. We have changed the descriptions of L368-374 as follows, and removed Fig. 6 and Fig. S10.

"In addition to aggravating pollutant accumulation, transported dust can affect the environment and pollutant concentrations in other ways. To quantitatively demonstrate the impacts of dust on various pollutants, we introduced growth rate in the comparative analysis (Supplementary Sect. S9). For convenience, we defined the comparison of the results of March 6 and 15, 2021, as precomparison (PRE), and we defined the comparison between March 15 and 22, 2021, as postcomparison (POST)."

Instead, we depict a relative difference in optical intensity between dusty day and clean days as Fig. 10. This Figure is used to describe the impacts of dust storm on light intensity, due to the light attenuation of dust and enhanced aerosols.

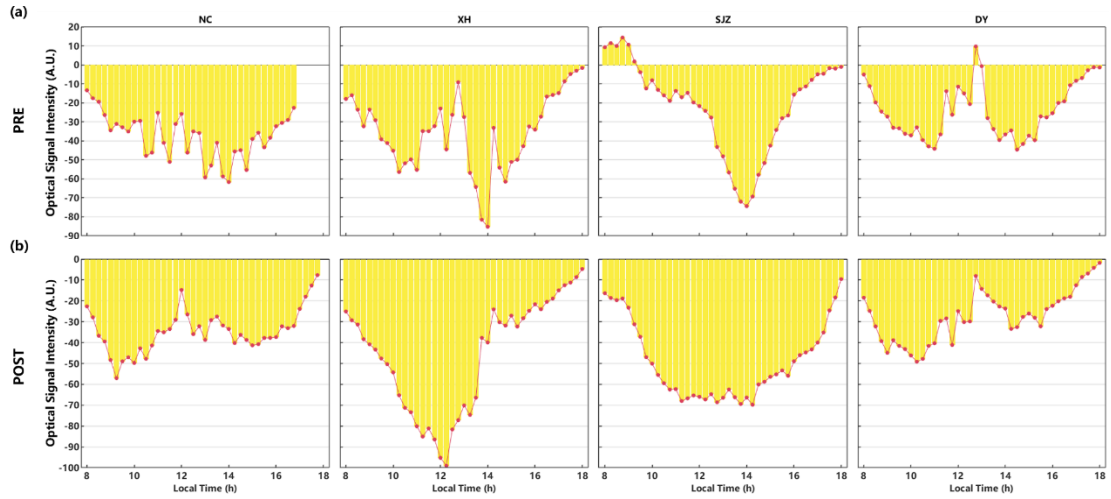


Fig. S10. The difference of optical signal intensities received by MAX-DOAS between dusty day and clean days. (a) PRE: Intensity (March, 15) – Intensity (March, 6); (b) POST: Intensity (March, 15) – Intensity (March, 22).

Finally, the manuscript appears to require substantial revision regarding of the usage of the English language. I have mentioned only a few in the technical corrections below.

Re: We let a language revision institution help us polish our language and correct mistakes.

Specific Comments:

L46: How does the transport of pollutants lead to the production and emission of pollutants? Please explain.

Re: Thank you for this comment. We have changed this sentence to avoid misunderstanding.

"transportation directly deteriorates the environment through the production and emission of a large number of pollutants" -> "transport carries large amounts of pollutants, directly deteriorating air quality."

L73: This sentence is not only too general, but also incorrect. Satellite remote sensing data is certainly extremely useful to monitor variations in the atmospheric composition (although with no or only limited vertical resolution in the troposphere).

Re: Thank you for this comment. We have changed our descriptions.

"Furthermore, satellite remote sensing data cannot be used to monitor the vertical features of and variations in the atmospheric composition." -> "However, because of their limited temporal and spatial resolutions, satellite data cannot be used for the continuous monitoring of a specific area (Bessho et al., 2016; Veefkind et al., 2012). It is difficult to characterize the vertical distribution of atmospheric composition using only satellite remote sensing or CNEMC data."

L76: I guess the statement "Large uncertainties remain in pollutant distribution estimation" only refers to model simulations. Please clarify.

Re: Yes, it only refers to model simulations. To avoid misunderstanding, we have changed this sentence.

"Large uncertainties remain in pollutant distribution estimation, primarily owing to the effects of emission inventories, meteorological fields, and hypothetical conditions (Huang et al., 2016; Xu et al., 2016; Zhang et al., 2017)." -> "However, considerable uncertainties remain in estimating pollutant distribution using model simulations, primarily owing to the effects of emission inventories, meteorological fields, and of assumptions made (Grell et al., 2005; Huang et al., 2016; Xu et al., 2016; Zhang et al.,

2017).”

L77: What do you mean with "hypothetical conditions"?

Re: Maybe using “assumptions” is better. As we all know, there are some basic model assumptions in simulation models (e.g., WRF/Chem, MM5/Chem), and the uncertainties caused by these assumptions cannot be ignored.

For example, Grell et al. (2005) indicated that the leaf temperature assignment assumption influenced O_3 statistics. In addition, other model components, such as surface layer parameterizations and boundary condition assumptions, contributed to model uncertainty.

L86: Describing DOAS as a "a cutting-edge and promising method" seems inappropriate. DOAS is a well established and well validated technique that has been applied for the measurement of atmospheric trace gases since decades.

Re: Thank you for this comment.

“The differential optical absorption spectroscopy (DOAS) technique (Platt and Stutz, 2008) is a cutting-edge and promising method for the quantitative analysis of many crucial atmospheric gases.” -> “The differential optical absorption spectroscopy (DOAS) technique (Platt and Stutz, 2008) is a well-established and reliable method for the quantitative analysis of many crucial atmospheric gases.”

L93: I think this statement is not correct, since LIDAR has a much better vertical resolution than MAX-DOAS (at least for aerosols).

Re: Thank you for this comment. We have amended this sentence.

“Compared with the above techniques, MAX-DOAS is high resolution and low cost, and its operation is automatic.” -> “Compared with the above techniques, MAX-DOAS does not require radiometric calibration and has many other advantages such as simple design, low power demand, possible automation, low cost, and minimal maintenance.”

L96: MAX-DOAS is a not a hyperspectral method since spectral information is only obtained from a single viewing direction at one time. It is also not clear why MAX-DOAS should be a stereoscopic technique.

Re: We have deleted this description to make the sentence more reasonable.

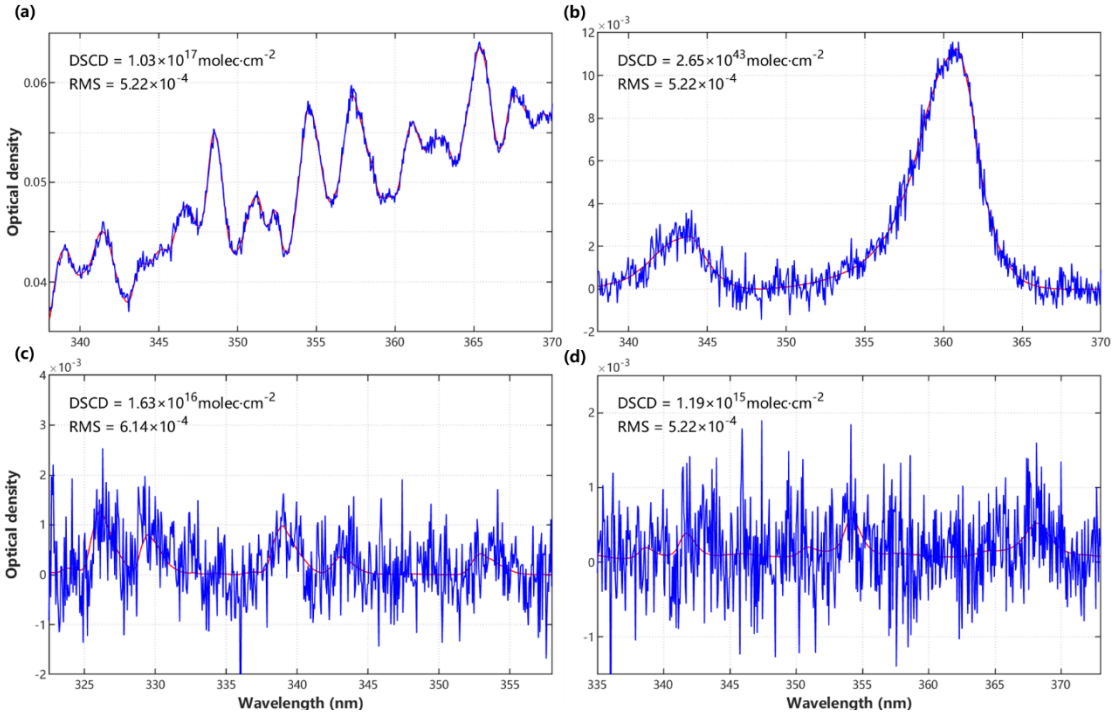
“a mature ground-based hyperspectral stereoscopic remote sensing network” -> “a mature ground-based remote sensing network”

Section 2.3: Please discuss the fit errors and **detection limits** for the retrieved species. The optical density of HCHO and HONO shown in figure S2 are very weak. Can these trace gases be detected reliably, and is the signal-to-noise ratio sufficient for a useful retrieval of the vertical distribution of HONO and HCHO?

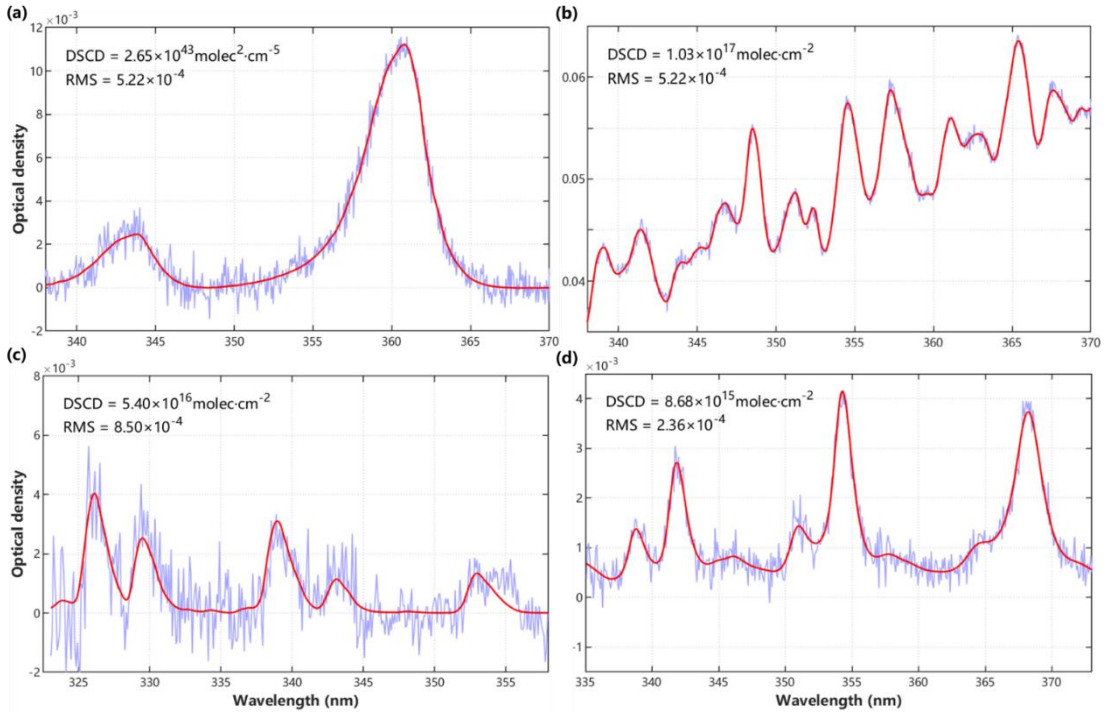
Re: We have supplemented the discussion about the fit errors and detection limits in Section. 2.3 as follows.

“We assumed two times the fitting error RMS as the DSCD detection limits (Wang et al., 2017; Lampel et al., 2015), which were 7×10^{41} ($molec^2 \cdot cm^{-5}$), 1.6×10^{15}, 3.6×10^{15}, and 5.8×10^{14} $molec \cdot cm^{-2}$ for O_4, NO_2, HCHO, and HONO, respectively.”

The results below can meet the RMS ($< 1 \times 10^{-3}$) and detection limit filtering criteria.



But we plan to use another HCHO and HONO result, and change the colors usage in this Figure as follows.



L140: Here it is not clear what you mean with "we calculated the ring spectrum as the measured spectrum, considering the contribution of the stratosphere to the DSCDs".

Re: “Furthermore, we calculated the ring spectrum as the measured spectrum, considering the contribution of the stratosphere to the DSCDs” -> “The Ring spectrum was added to the fitting settings to remove the influence of the stratosphere on the DSCDs.”

Section 2.4: Given that the signal-to-noise ratio apparent in Figure S2 seems to be very low, I am surprised that the smoothing and noise error components of the HCHO profiles are similar to those of the NO2 profile. It is not clear what you mean with "algorighmic error". Is this error due to inaccurate model parameters b or due to general incapabilities of the forward model to realistically represent the underlying physics?

Re: Actually, the HCHO smoothing and noise error is larger than the NO_2 result, especially for near-surface value (HCHO:42%, NO_2:11%), which agrees with the SCD retrieved results.

The descriptions about "algorithmic error" might be a little confusing. We have changed the demonstrations in the main text as follows and further elaborate it in Supplementary Sect. S3.

Main text: "Algorithmic error (i.e., the difference between the measured and modeled DSCDs) arises from an imperfect minimum of the cost function. This error is a function of the viewing angle. However, it is difficult to assign discrepancies between the measured and modeled DSCDs at each profile altitude. Therefore, the algorithm error on the near-surface values and column densities cannot be realistically estimated. Given that measurements at 1° and 30° elevation angles are sensitive to the lower and upper air layers, respectively, the average relative differences between the measured and modeled DSCDs for a 1 and 30° elevation angles can be used to estimate the algorithm errors on the near-surface values and column densities, respectively (Wagner et al., 2004). Considering its trivial role in the total error budget, we estimated these errors on the near-surface values and the column densities at 4 and 8 % for aerosols, 3 and 11 % for NO_2, and 4 and 11 % for HCHO, respectively, according to Wang et al. (2017)."

Supplementary Sect. S3: "The algorithm error is the discrepancy between the measured ($\boldsymbol{y}$) and modelled DSCDs ($F(\boldsymbol{x},\boldsymbol{b})$), which is mainly caused by an imperfect minimum of the cost function (χ^2) in Eq. s3. This error is a function of the viewing angle. Due to the difficulty of assigning this error to each altitude of profile, the algorithm errors on the near-surface values and column densities are usually estimated by calculating the average relative differences between the measured and modeled DSCDs at the minimum and maximum elevation angle (except 90°), respectively (Wagner et al., 2004).

$$\chi^2 = (\mathbf{y} - F(\mathbf{x},\mathbf{b}))^T \mathbf{S}_\varepsilon^{-1} (\mathbf{y} - F(\mathbf{x},\mathbf{b})) + (\mathbf{x} - \mathbf{x}_a)^T \mathbf{S}_a^{-1} (\mathbf{x} - \mathbf{x}_a) \quad \text{(s3)}$$

where $F(\boldsymbol{x},\boldsymbol{b})$ is the forward model; $\boldsymbol{b}$ represents the meteorological parameters; $\boldsymbol{y}$ is the measured DSCDs; $\boldsymbol{x}_a$ is the a priori vector that serves as an additional constraint; $\boldsymbol{x}$ is the state vector."

L208: Explain why the wind-speed in north-easterly direction (and not in any other direction) is of relevance here.

Re: Thank you for this comment. We have supplemented explanations at the start of Section 2.5.

"Given the major role of pollutant transports in the JJJ region, hourly transport fluxes of each layer ($Flux_i$) and column transport fluxes ($Flux_c$) at each station were

calculated to illustrate the dynamic transport process of pollutants along southwest-northeast pathway." -> "Owing to the semi-basin topography, southwesterly or southerly winds play a dominant role in pollutant transport in the JJJ region. In this study, we mainly focused on pollutant transport in the southwest-northeast direction, and thus selected four different stations along this pathway, namely, Shijiazhuang (SJZ), Wangdu (WD), Nancheng (NC), and Chinese Academy of Meteorological Sciences (CAMS) (Fig. 1). We calculated the hourly transport fluxes of each layer (F_i) and column transport fluxes (F_c) at each station to illustrate the dynamic transport process of pollutants along the southwest-northeast pathway."

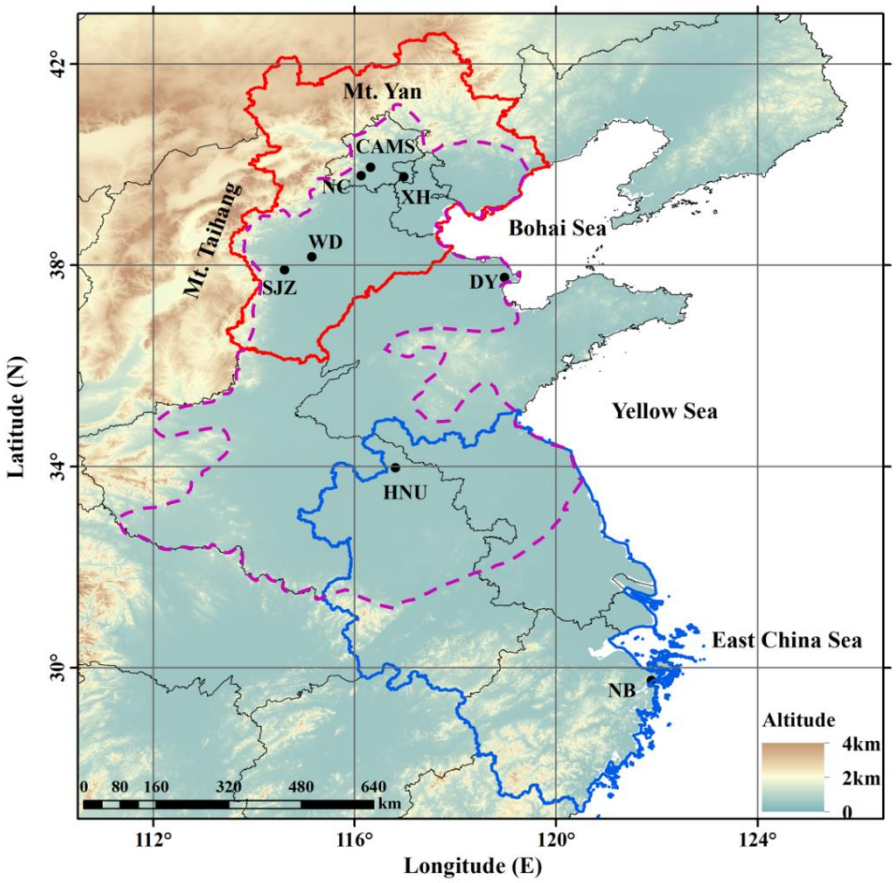


Figure 2: Converting the in situ NO2 from μ g/m3 to ppb would allow for a much better quantitative assessment of the agreement between both datasets.

Re: We have converted unit of NO2 from μ g/m3 to ppb using the following formula.

$$C(ppb) = \frac{X(\mu g/m^3)\cdot V_m(L/mol)}{M_g(g/mol)}$$

$$V_m(L/mol) = \frac{R(J/(mol\cdot K))\bullet T(K)}{P(kpa)}$$

In addition, to lessen the effects of occasional extreme conditions, we filtered the "abnormal values" of MAX-DOAS and in situ measurements before comparison. This filtering helped improve the correlations from 0.615 to 0.752, and 0.671 to 0.74, for

aerosol and NO_2, respectively.

The filtering procedure is elaborated in Supplementary Sect. S8 as follows.

"**Section S8. The abnormal values definition and filtering**

For lessening the impacts of "abnormal value" caused by occasional extreme conditions, we needed to adopt a method to seek out the abnormal values and filter them out. In a series of data, we firstly found the first quartile (Q1), median (Q2), and the third quartile (Q3), which are the 25th, 50th and 75th percentile of all values from small to large, respectively. The difference between Q1 and Q3 is called interquartile range (IQR) (i.e., IQR = Q3-Q1). The upper limit (L_{upper}) and lower limit L_{lower} were defined as Q3 plus IQR, and Q1 minus IQR, respectively (i.e., L_{upper}=Q3+IQR, L_{lower}=Q1-IQR). The values larger than L_{upper} or lower than L_{lower} were defined as abnormal values, and discarded. After filtering the data, the correlation had increased from 0.615 to 0.752, and 0.671 to 0.74, for aerosol and NO_2, respectively.

"

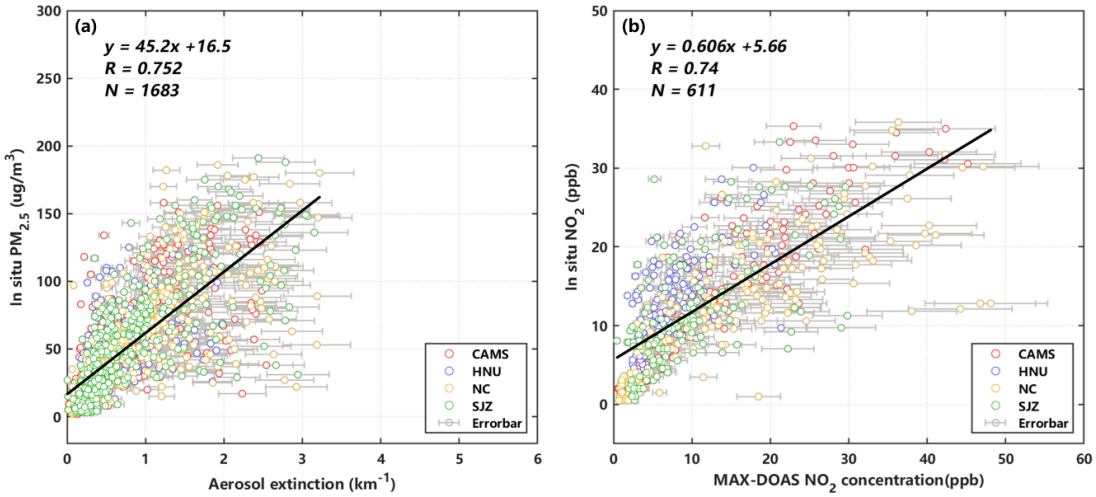
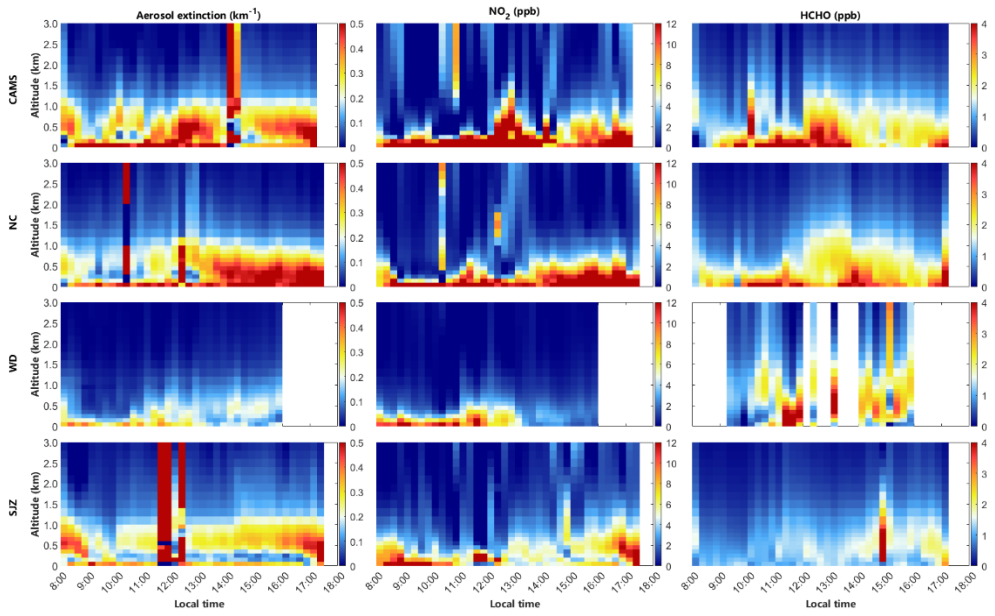


Figure 3: Why are there so many missing profiles? Is this due to outages of the instrument or has the profile retrieval failed in these cases?

Re: On one hand, outages of the instrument result in some periods of failure, and make some stations lack SCDs during the certain period. For example, the vertical profiles are lacked after 16:00 at the WD station, which is mainly attributed to measurement failure. On the other hand, some results cannot meet the selection criteria (DOF > 1.0, relative error < 50%), and are filtered out. If we don't filter the data, there will be more profiles, but many results are obviously unreasonable (as follows). For instance, there are >0.5 km^{-1} AECs at each altitude during 11:00-12:00 at the SJZ station.



L270ff: I feel that the description of the temporal and vertical distribution of aerosols at the different stations is not representing the overall picture appropriately. For example, it is stated that there is a "subtle increase" in aerosols above NC around 12:00, but it is not mentioned that this is just the onset of the presence of a strong aerosol layer throughout the afternoon. The finding that a persistent and elevated aerosol layer is first present at SJZ, and later at NC and CAMS, is not explicitly discussed. Are the times at which the aerosol layer reaches the different locations in agreement with the transport times from station to station as estimated from the wind speed? This would give further evidence that long-range transport has indeed occurred. What could be the reason for the much lower AECs at WD than at the other stations?

Re: Thank you for this comment. As you said, we didn't make clear distinctions between (1) conclusions evidently inferred from the measurements, (2) findings from other studies that support the measurements and (3) hypotheses based on the observations. Only with existing data, it is not convincing to discuss whether the distance fits together with the wind speed and the time distance. This is because the distance among SJZ, WD, and NC is too long (> 40 km), and wind speeds would experience many uncertain changes during this process. Maybe these arguments should be considered as hypotheses based on the observations instead of conclusions evidently inferred from the measurements. Given that the main points of Section 3.1 are to discuss the MTL of various pollutants, we decide to delete such descriptions.

To emphasize the main points, we have changed structure of Section 3.1. For Fig.3, we mainly discuss spatiotemporal distributions of aerosol, NO_2, and HCHO at different stations during this regional transport as follows. And the reason for the much lower AECs at WD than at the other stations is also contained in the following descriptions.

"Figure 3 presents the temporal variations in the vertical distributions of aerosols, NO_2, and HCHO during this regional transport. At the CAMS and NC stations, the aerosol, NO_2, and HCHO concentrations were consistently high near the surface, primarily because of the heavy traffic flow and dense factory emissions in Beijing (Zhang et al.,

2016; Li et al., 2017). Previous studies have suggested that urban air pollution in Beijing is dominated by a combination of coal burning and vehicle emissions, which results in severe particulate pollution (Wang and Hao, 2012; Wu et al., 2011). At SJZ, the NO_2 concentration was high (~ 12 ppb) in the morning and late afternoon, whereas the concentration was lowest (~ 6 ppb) near noon, which is explained by the morning and evening rush hour. Comparatively, the overall AEC and NO_2 levels were relatively low at the WD station, whereas a continuous high-value HCHO distribution (> 2 ppb) occurred at 0-1500 m between 11:00 and 16:00. This occurred because the WD station is located in a farm field with less traffic flow and high vegetation coverage; therefore, large amounts of HCHO are directly emitted by biogenic sources and secondarily produced by natural and anthropogenic volatile organic compound (VOC) photolysis (Wang et al., 2016; Wu et al., 2017a)."

L297: How exactly do NO2 and HCHO enhance the AEC?

Re: Thank you for this comment. We speculated that NO_2 and HCHO enhance the AEC through secondary aerosol generations. As you mentioned in General Comments, we lacked direct evidence from the measurements that support this finding. Considering that this is not a conclusion evidently inferred from the measurements, we have deleted these descriptions.

L300ff: How do you know that secondary aerosols were generated? To my knowledge, this cannot be inferred from MAX-DOAS measurements. I cannot find any evidence for your statement that secondary aerosol generation is always accompanied the regional transport process. Does that come from model calculations or other measurements? If so, please explain in detail. Is NO2 really the main precursor for organic aerosols? Please cite relevant publications that support this statement.

Re: Thank you for this comment. Secondary aerosols generation is just one of our speculations. We didn't conduct any model calculations or other measurements here. Considering that it is less associated with the main points of Section 3.1, we have deleted these demonstrations.

L277: It appears from Fig. 3 that the decrease in MTL of aerosols at NC already occured at 14:00, not 16:00. I do not think that the decrease in the aerosol layer height is related to the formation of a nocturnal boundary layer, which is formed much later right before sunset, and is initially very shallow. Aerosols present aloft would reside in the residual layer above the nocturnal surface layer. It seems much more likely that the increase in aerosol layer width is instead caused by increased vertical mixing due to a heat up of the surface in the course of the day.

Re: Thank you for this suggestion. We have changed our demonstrations here.

"We observed a similar phenomenon at NC around 16:00 and at CAMS after 17:00. Surface aerosol accumulation is closely linked with the collapse of the mixing layer and the formation of a stable nocturnal boundary layer (Ding et al., 2008; Ran et al., 2016), triggering a descending tendency in the MTL." -> "In the late afternoon, aerosols gradually accumulated towards the surface, and triggered a variation in the distribution of F_i. After 16:00, the shift in the high-AEC air mass caused the transport fluxes in the lower layers (100–200 m) to increase to > 1.1 and ~ 2 $km^{-1} \cdot m \cdot s^{-1}$ for the CAMS and SJZ stations, respectively. Surface aerosol accumulation is closely linked to the collapse

of the mixing layer and formation of a stable nocturnal boundary layer (Ding et al., 2008; Ran et al., 2016). Remarkably, high-altitude aerosol air masses began to mix with near-surface aerosols after 14:00 at the NC station (Fig. 3), triggering a variation in the MTL (Fig. 4). This might be explained by enhanced vertical mixing due to the heating of the surface during the course of the day (Castellanos et al., 2011; Wang et al., 2019). Generally, the MTL of aerosols was situated at 400–800 m during the daytime, where variations in the boundary layer and increased vertical mixing can influence the MTL."

L285ff: According to Eq. 3, shouldn't the unit for trace gas flux be ppb·m·s^{-1}, and for aerosol extinction km^{-1}·m·s^{-1}?

Re: Yes. It's our fault to make descriptions jump all the time between Fig. 3 and Fig. 4, without inserting a Figure number here. L285ff is the description about Fig. 3. To avoid misunderstanding and make the paragraphs more understandable, we have changed the structure of Section 3.1. Now, we describe Fig. 3 firstly, and then mainly demonstrate Fig. 4 as follows.

"Figure 3 presents the temporal variations in the vertical distributions of aerosols, NO_2, and HCHO during this regional transport. At the CAMS and NC stations, the aerosol, NO_2, and HCHO concentrations were consistently high near the surface, primarily because of the heavy traffic flow and dense factory emissions in Beijing (Zhang et al., 2016; Li et al., 2017). Previous studies have suggested that urban air pollution in Beijing is dominated by a combination of coal burning and vehicle emissions, which results in severe particulate pollution (Wang and Hao, 2012; Wu et al., 2011). At SJZ, the NO_2 concentration was high (~ 12 ppb) in the morning and late afternoon, whereas the concentration was lowest (~ 6 ppb) near noon, which is explained by the morning and evening rush hour. Comparatively, the overall AEC and NO_2 levels were relatively low at the WD station, whereas a continuous high-value HCHO distribution (> 2 ppb) occurred at 0-1500 m between 11:00 and 16:00. This occurred because the WD station is located in a farm field with less traffic flow and high vegetation coverage; therefore, large amounts of HCHO are directly emitted by biogenic sources and secondarily produced by natural and anthropogenic volatile organic compound (VOC) photolysis (Wang et al., 2016; Wu et al., 2017a).

Nevertheless, Fig. 3 cannot reveal the exact layers in which the main transport phenomena occur. For instance, at the CAMS station, the AEC at the surface and upper layers both reached ~ 0.5 km^{-1} around noon, making it difficult to determine the layer in which transport was more obvious. To further demonstrate the dynamic transport process of different pollutants, we calculated the hourly F_i and F_c, and defined the MTL. As shown in Fig. 4, a positive F_i indicates that all transport flux projections in the southwest-northeast pathway are from southwest to northeast at the four stations. The MTLs of aerosols, HCHO, and NO_2 exhibited different spatiotemporal characteristics. Although surface and high-altitude (400–800 m) AECs both remained at a relatively high level (> 0.3 km^{-1}) at CAMS during 12:00-17:00 (Fig. 3), there was a large discrepancy between their corresponding F_i values (Fig. 4). The aerosol near-surface F_i was ~ 1 km^{-1}·m·s^{-1} after 12:00, while F_i in layers of 400–800 m all exceeded 1.2 km^{-1}·m·s^{-1}, and even reached ~ 2 km^{-1}·m·s^{-1} around 12:00. At the SJZ station, the AECs at surface and 300–1000 m layer mostly ranged from 0.3 to 0.4 km^{-1},

especially after 10:00 (Fig. 3). However, the MTLs of aerosols were mostly at 400–800 m throughout the day, with many transport fluxes in those layers even reaching ~ 2 $km^{-1} \cdot m \cdot s^{-1}$ (Fig. 4). At the WD station, the highest F_i also tended to occur at high layers (400–800 m), with maximum F_i exceeding 1.7 $km^{-1} \cdot m \cdot s^{-1}$ at 400–500 m at 15:00. This suggested that aerosol transport occurred mainly in the upper layers. In the late afternoon, aerosols gradually accumulated towards the surface, and triggered a variation in the distribution of F_i. After 16:00, the shift in the high-AEC air mass caused the transport fluxes in the lower layers (100–200 m) to increase to > 1.1 and ~ 2 $km^{-1} \cdot m \cdot s^{-1}$ for the CAMS and SJZ stations, respectively. Surface aerosol accumulation is closely linked to the collapse of the mixing layer and formation of a stable nocturnal boundary layer (Ding et al., 2008; Ran et al., 2016). Remarkably, high-altitude aerosol air masses began to mix with near-surface aerosols after 14:00 at the NC station (Fig. 3), triggering a variation in the MTL (Fig. 4). This might be explained by enhanced vertical mixing due to the heating of the surface during the course of the day (Castellanos et al., 2011; Wang et al., 2019). Generally, the MTL of aerosols was situated at 400–800 m during the daytime, where variations in the boundary layer and increased vertical mixing can influence the MTL. In contrast to aerosols, we found that a high-value NO_2 F_i frequently occurred in the 0–400 m layer. Except that F_i reached the highest level of ~ 50 $ppb \cdot m \cdot s^{-1}$ in the 400–600 m layer at 16:00 at the SJZ station, the other highest F_i all occurred below 400 m at any station and at any time. This indicated that the MTL of NO_2 was 0–400 m. Near-surface NO_2 emission sources (e.g., vehicle and factory emissions) might be the main reason for this phenomenon. Compared with aerosols and NO_2, we found that high-value HCHO F_i extended to higher altitudes. Taking CAMS as an example, we found the strongest HCHO F_i constantly emerging at 1000–1200 m from 8:00 to 13:00, and averaging 9.18 $ppb \cdot m \cdot s^{-1}$. During the same period, surface HCHO F_i only averaged 6.44 $ppb \cdot m \cdot s^{-1}$. However, at the CAMS station, the surface HCHO concentration was much higher than that of the 1000–1200 m layer between 8:00 and 13:00 (Fig. 3), proving that high-altitude transport contributed more to overall HCHO transport. After 10:00, we found that the highest HCHO F_i gradually increased from ~ 8 to ~ 20 $ppb \cdot m \cdot s^{-1}$ at WD, with the MTL of HCHO ranging from 400 to 1000 m. At station SJZ, the strongest HCHO F_i increased from ~ 10 to ~ 16 $ppb \cdot m \cdot s^{-1}$ during 11:00–17:00, with the highest transport fluxes occurring mostly at 400–800 m. These findings indicated that the MTL of HCHO was mainly 400–1200 m. The sharp variation in the MTL at the NC station might be caused by atmospheric vertical mixing (Castellanos et al., 2011; Wang et al., 2019). As shown in Fig. 3, high HCHO concentrations tend to appear at higher altitudes than those of aerosols and NO_2. A possible explanation is that the precursor compounds of HCHO are transported to higher layers and converted into HCHO through photochemical reactions, resulting in elevated HCHO concentrations at higher altitudes (Kumar et al., 2020). Furthermore, strong high-altitude winds were more conducive to HCHO transport (Fig. S5), which further increased the corresponding transport flux. Notably, HCHO F_i was enhanced around noon because the increased solar radiation promotes the secondary generation of HCHO. Long-term observations have revealed that secondary HCHO formation through VOCs photolysis plays a significant role in

Beijing (Liu et al., 2020; Zhu et al., 2018)."

L323: What kind of satellite results are your referring to? This should be explained in the main text, but is not even clear from the caption of Fig. S5.

Re: "The satellite" referred to the Himawari-8, which can monitor the spatial distribution of aerosols and dust.

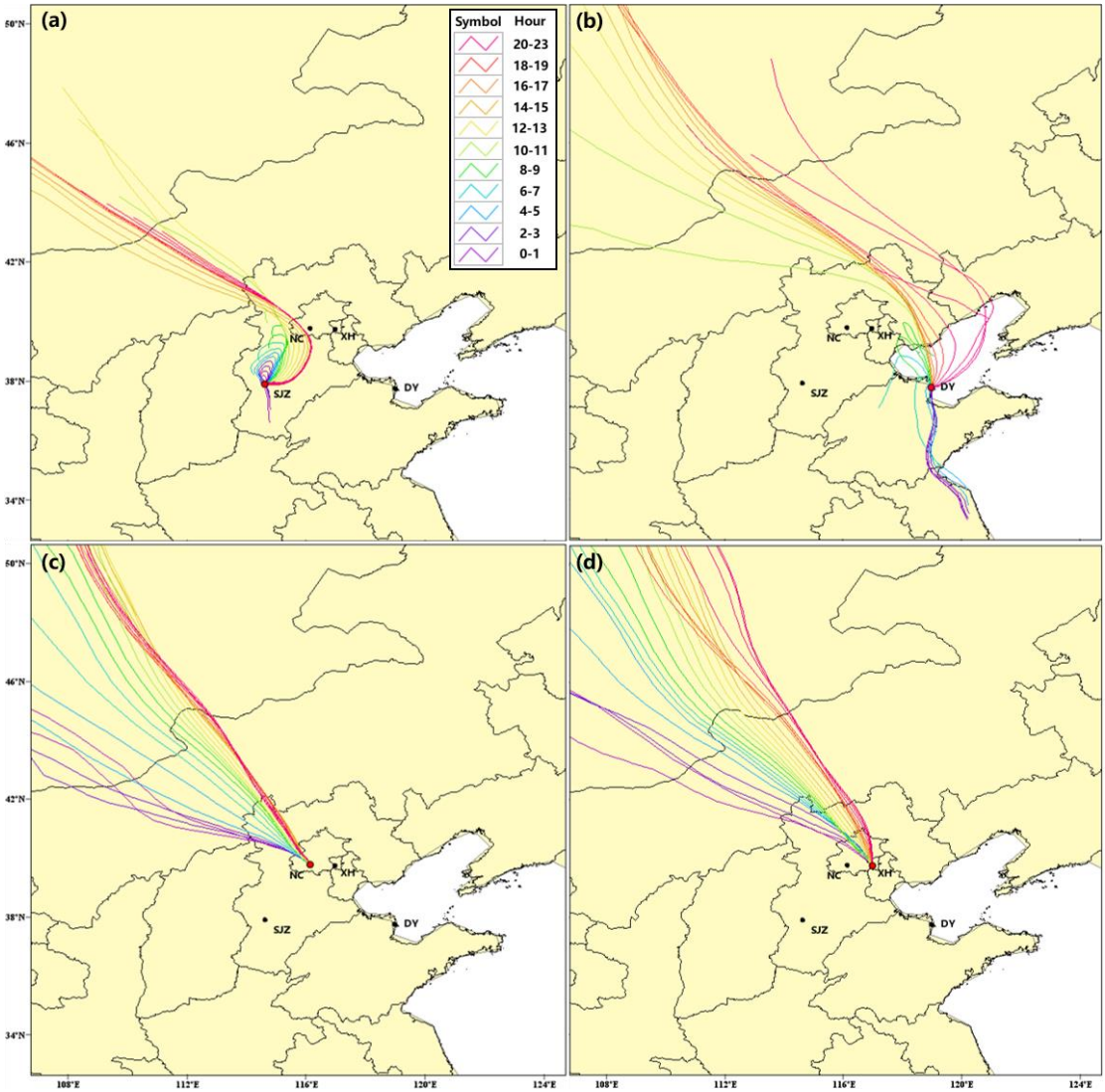
To make it clearer, we have changed some words.

Main text: "The satellite results…" -> "The Himawari-8 observations…"

Supplementary materials: "**Fig. S5.** A severe dust storm invaded northern China at (a) 8:00 and (b) 14:00 on March 15, 2021." -> "**Fig. S8.** The Himawari-8 observations: a severe dust storm invaded northern China at (a) 8:00 and (b) 14:00 on March 15, 2021. The dashed black contour line indicates the NCP region."

Fig. S6: It is not clear what is shown here. Are these trajectories at different times or at different heights? The trajectories should be colour-coded for different heights/times.

Re: These trajectories are at different times. We have color-coded the trajectories for different times and added description information as follows.



"Fig. S6. The 24-h backward trajectory results of (a) SJZ, (b) DY, (c) NC, and (d) XH on March 15, 2021 by means of the HYSPLIT model." **-> "Fig. S9.** The 24-h backward

trajectory results of (a) SJZ, (b) DY, (c) NC, and (d) XH from 00:00 to 23:00 on March 15, 2021 by means of the HYSPLIT model. The altitude of the receptor site was set to the 100 m above ground level.
”

Technical Corrections

L50: "driven by the southwest wind" -> "driven by south-westerly winds"
L66: "pollutant concentrations monitoring" -> "pollutant concentrations monitored"
L67: "Characterize" -> "Characterizing"
L74: "The chemical transport model" -> "Chemical transport models"
L75: "pollutant distribution" -> "pollutant distributions"; "is" -> "are"
Section 2: The title "Method and methodology" is a tautology. Use either "Methods" or "Methodology"
Re: Thank you for these corrections. We have followed the suggestions above and corrected all of them.
L124: "We operated a commercial MAX-DOAS instrument" -> "We operated seven (?) commercial MAX-DOAS instruments"
Re: “We operated a commercial MAX-DOAS instrument (Airyx, Heidelberg, Germany)” -> “We operated eight commercial MAX-DOAS instruments (Airyx SkySpec-1D, Heidelberg, Germany)”
L140: Please explain what SCDs are (integrated concentrations along the light path).
Re: We have added a sentence before to explain SCDs.
“Spectral analysis derives the slant column densities (SCDs), i.e., the integrated concentration along the light path.”
Section 2.6: According to the ACP guidelines, all variables should be named according to the IUPAC conventions, with all variables being named using only a single lower-case letter. For example, in Equation (2) the expression va would be by convention interpreted as v·a, which is not what you mean here. I would suggest to use u and v for the meridional and zonal wind, respectively, and to replace Fluxc with Fc, and WSi with wi.
Re: Thank you for this suggestion. We have corrected these problems.
L221: Do you mean "layer with highest transport"?
Re: “the layer with high transport flux” -> “the layer with the highest transport flux”
L223: "discrepancy" -> "differences"
L257: Two times "that".
Re: Thank you for these corrections. We have followed the suggestions above and corrected all of them.
L258: "continuously" -> "homogeneously"
Re: “According to the TROPOMI results, we found that that NO_2 was continuously distributed between SJZ and WD, whereas a HCHO distribution belt connected NC with CAMS on February 5, 2021 (Fig. S2).” -> “The TROPOMI results indicated that NO_2 was homogeneously distributed between SJZ and WD and that, on February 5, 2021, the HCHO distribution belt connected NC with CAMS (Fig. S4).”

References

Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frost, G., Skamarock, W. C., and Eder, B.: Fully coupled “online” chemistry within the WRF model, Atmos Environ, 39, 6957-6975, 10.1016/j.atmosenv.2005.04.027, 2005.