Comment on egusphere-2022-653
Anonymous Referee #3


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.

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.
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.

**Specific Comments**

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

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).

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

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

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.

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

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.

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?
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".

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 "algorithmic error". Is this error due to inaccurate model parameters or due to general incapabilities of the forward model to realistically represent the underlying physics?

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

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.

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?

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?

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

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.

L277: It appears from Fig. 3 that the decrease in MTL of aerosols at NC already occurred 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.

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?

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.

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.

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"

L124: "We operated a commercial MAX-DOAS instrument" -> "We operated seven (?) commercial MAX-DOAS instruments"
L140: Please explain what SCDs are (integrated concentrations 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 $v \cdot a$, 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 $Flux_c$ with $F_{ci}$ and $WS_i$ with $w_i$.

L221: Do you mean "layer with highest transport"?

L223: "discrepancy" -> "differences"

L257: Two times "that".

L258: "continuously" -> "homogeneously"