Referee #1

The manuscript developed an updated NO_x emission inventory in East Asia (constraint with GEMS) and analyze source-receptor relationship between China and S. Korea during 2022 winter-spring transition. The manuscript is well-written. The logic is clear, method being sound, and results are convincing. I only find few minor limitations need to address as follows:

Comment	1) line 28, it is better to provide the lifetime of NO_x in days, cause the transport across yellow sea can also be short. 2) line 32. it is better to add some reference for long-range transport studies in Korea domain. there are many published during KORUS-AQ campaign, and you can compare your results here.
Response	Thank you for pointing these out. We acknowledge the need to further elaborate on the NO_x lifetime and to provide additional context. We have revised the manuscript with additional details and references.
Changes made	L28-38: NO_x has a relatively short atmospheric lifetime, typically ranging from a few hours … to several days … contributing to both local and transboundary air quality challenges.

Comment	3) line 72, need to cite paper for Korea-China transport studies for example: Eck et al, 2020 "Influence of cloud, fog, and high relative humidity during pollution transport events in South Korea: Aerosol properties and $PM_{2.5}$ variability" Jordan et al, 2020 "Investigation of factors controlling $PM_{2.5}$ variability across the South Korean Peninsula during KORUS-AQ" Nault et al, 2018, "Secondary organic aerosol production from local emissions dominates the organic aerosol budget over Seoul, South Korea, during KORUS-AQ" Lee et al, 2020 "Sensitivity of simulated $PM_{2.5}$ concentrations over northeast Asia to different secondary organic aerosol modules during the KORUS-AQ campaign" Choi et al, 2019. "Impacts of local vs. trans-boundary emissions from different sectors on $PM_{2.5}$ exposure in South Korea during the KORUS-AQ campaign" Tang et al, 2023 "WRF-Chem quantification of transport events and emissions sensitivity in Korea during KORUS-AQ" Bae et al, 2020 "Long-range transport influence on key chemical components of $PM_{2.5}$ in the Seoul Metropolitan Area, South Korea, during the years 2012-2016"
Response	Thanks for your suggestion and for sharing relevant references. We have incorporated these citations into our manuscript to enhance the introduction accordingly.
Changes made	L71-83: “Bae et al., (2020) investigated the influence of NO_x … Similarly, Lee et al.’s study (2020) demonstrated that the long-range transport … Tang et al. (2023) also assessed the contributions of … 51% and 70% of the total $PM_{2.5}$ mass, respectively.” L127-135: “Beyond these satellite-based studies … Choi et al. (2019)’s study utilized in-situ observation data … Despite these efforts,”

Comment	4) line 90 to 120. need to mention Dr. Woo's KORUS-AQ v5 emission and also recent emission used for ASIA-AQ. those are state-of-art emissions widely used in research community and require an acknowledgement. 8) line 162, why do not use Dr. Woo's emission prepared for ASIA-AQ? it should be more accurate and widely used. if not, please address the reason in the manuscript.
Response	Thank you for the suggestion. We are aware of the exceptional utility and maturity of Dr. Woo's group's emissions inventory, having extensively utilized it in our previous studies. We chose to elaborate on this in the methodology section rather than in the introduction, where the focus is on the broader limitations of existing regarding the inventories (we do not intend to challenge the KORUS-AQ inventory with our 'top-down' estimates). Nevertheless, we have acknowledged the KORUS-AQ inventory and clarified our rationale for selecting the EDGAR inventory for our study.
Changes made	L211-219: "It is noteworthy that, unlike our previous study (Park et al., 2023) … was delicately built upon a combination of multiple regional surveys, providing comprehensive representations of emissions patterns across Asia … to avoid introducing additional uncertainties associated with the temporal allocation of monthly emissions into hourly emissions."

Comment	5) line 120, please provide evidence than long-range transport only occur within PBL or rephrase.
Response	Thanks for pointing this out and we agree that the original description could mislead readers, as transport does not occur exclusively within the PBL. We have rephrased the relevant line and included additional clarifications in the methodology section.
Changes made	L143-145: "Then, using CMAQ-ISAM, we quantified the local and transboundary contributions of NO_x emissions to NO_y concentrations ~~within the planetary boundary layer (PBL)~~ across five major NO_x source regions of East Asia during the period from January to May 2022. L191-195: "However, it is important to acknowledge that substantial pollutant transport also occurs in the free troposphere beyond the PBL, where stronger winds facilitate long-range movement of pollutants. Our study specifically focuses on the PBL to assess cross-regional pollutant behaviors, as this layer directly influences surface air quality, the modeled estimates of which can be evaluated with station measurements (detailed in Section 2.5), and human health associated."

Comment	6) line 130, need a brief description of what schemes, initial and boundary conditions you used here. I saw you sum them up in supplemental materials, but it is useful to introduce them in the manuscript as well.
Response	Thank you for your suggestion. We agree that including those details would improve the manuscript. We have accordingly enriched the methodology section, summarizing key configurations.
Changes made	L156-162: "We used the Morrison two-moment scheme for microphysics … from the National Centers for Environmental Prediction (NCEP) FNL operational model global tropospheric analysis." L175-180: "We used the YAMO scheme and the WRF omega formula … static boundary conditions during the entire simulation period."

Comment	7) Figure 1. do you have plan to account N. Korea emission impact, it is much close to S. Korea and should have more close impact on S. Korea air quality? if not, please rephrase and explain reason.
Response	Thank you for your insightful concern. We recognize North Korea's proximity to South Korea and acknowledge the potential for its emissions to influence air quality in the region. Accurately accounting for North Korea's emissions poses is challenging due to the lack of reliable input data. Global emissions inventories like EDGAR, which rely heavily on accurate energy usage data to estimate pollutant emissions, have considerable uncertainty in regions where such data is unavailable or incomplete. This rationale has been clarified in the manuscript.
Changes made	L184-185: "North Korea, despite its close proximity to these regions and the potential impact of its emissions on neighboring regions' air quality … the lack of reliable input data for North Korea made it impractical to include as a separate source region in this study."

Comment	4) line 90 to 120. need to mention Dr. Woo's KORUS-AQ v5 emission and also recent emission used for ASIA-AQ. those are state-of-art emissions widely used in research community and require an acknowledgement. 8) line 162, why do not use Dr. Woo's emission prepared for ASIA-AQ? it should be more accurate and widely used. if not, please address the reason in the manuscript.
Response	Thank you for the suggestion. We are aware of the exceptional utility and maturity of Dr. Woo's group's emissions inventory, having extensively utilized it in our previous studies. We chose to elaborate on this aspect in the methodology section rather than in the introduction, where the focus is on the broader limitations of existing inventories (we do not aim to challenge the KORUS-AQ inventory with our top-down estimates). We have acknowledged the contributions of the KORUS-AQ inventory and explained our rationale for selecting the EDGAR inventory for this study.
Changes made	L211-219: "It is noteworthy that, unlike our previous study … was delicately built upon a combination of multiple regional surveys … to avoid introducing additional uncertainties associated with the temporal allocation of monthly emissions into hourly emissions."

Comment	9) line 208, "assume a linear relationship between NO_2 column and emission". linear relationship is not real. please provide your reason why assuming linear? or any reference to prove the method.
Response	Thank you for raising this point. We agree that further clarification is necessary regarding the assumption we made. The assumption of a linear relationship between NO_x emissions and NO_2 concentrations is a widely adopted simplification in inverse modeling studies (listed in the manuscript). This approach is based on the understanding that satellite-observed NO_2 columns are primarily influenced by recent, localized NO_x emissions due to the short lifetime of NO_2. Under these conditions, changes in NO_x emissions are likely to result in proportional changes in NO_2 column densities, making the linearity assumption a practical method for constraining emissions. This assumption also simplifies the inversion process by directly linking observed NO_2 columns to emissions, avoiding the need for computationally intensive modeling of nonlinear chemical reactions and transport processes (in our study, the use of CMAQ DDM-3D partially accounts for the nonlinearity). We have clarified the rationale for this approach in the manuscript accordingly.
Changes made	L272-276: "This assumption, widely adopted in earlier inverse modeling studies… avoiding the need for computationally intensive modeling of nonlinear processes."

Comment	10) line 212, nighttime NO_2 chemistry is very important for NO_x distribution, giving that GEMS has no nighttime observation to constrain. please provide expected limitation of your study that ignore nighttime NO_x chemistry.
Response	Thank you for the insightful suggestion. We agree that this limitation should be clarified more explicitly in the manuscript. We have added the relevant explanation in the manuscript accordingly.
Changes made	L303-305: "However, a limitation of this approach is that it does not improve the representation of nighttime emissions, leaving these unadjusted due to the absence of observational constraints. Consequently, any model biases associated with nighttime emissions remain unresolved."

Comment	11) figure 2. when comparing GEMS NO_2 column with WRF-CMAQ model results, it is required that GEMS NO_2 is covered to use vertical profile from the same model. The default one for GEMS is NOT WRF-CMAQ. please provide how you do the conversion?
Response	Thanks for pointing this out. We have elaborated further on how we made the direct comparison between the GEMS's and CMAQ's NO_2 columns in the manuscript.
Changes made	L250-264: "including the averaging kernel, cloud fraction, data quality flags, … model-derived variables from the GEMS Level 2 data, including tropospheric and stratospheric air mass factors (AMF), a priori tropospheric NO_2 profile, and tropospheric pressure profile from the WRF model coupled with Chemistry (WRF-Chem) 3.9.1 (NIER, 2020). … excluded the cloudy scenes which could lead to inaccurate AMF references".

Referee #2

In this manuscript, the authors report on a study investigating the contribution of transboundary transport to NO_y levels in the Republic of Korea and China for January to May 2021. Using the CMAQ-ISAM, they first update the NO_x emission inventory used by inverting GEMS NO_2 observations. They then separate the region of interest into 4 areas and use tagged NO_y in the model to evaluate the contribution of different source areas to NO_y levels in East Asia during the winter spring transition.

The manuscript discusses an interesting topic. It is well structured and clearly written, and the methods and conclusions appear valid. However, there are several critical aspects which need to be addressed and clarified before the manuscript can be accepted for publication.

Major comments:

Comment	In the title and in other parts of the manuscript, the authors talk about "nitrogen loading". When I first read this, I expected that this would include ammonia and particulate nitrates, but this is not the case. Please use more precise wording to make clear what exactly is covered by this study.
Response	We agree that our wording "nitrogen loading" may mislead readers into expecting a broader scope, including species like ammonia and nitrate aerosols, which are not part of this study. We have revised the title and manuscript for clarity, accordingly.
Changes made	Title: Local and transboundary contributions to NO_y loadings across East Asia … during the winter-spring transition L8: "… to reactive nitrogen species (NO_y) loadings across East Asia …" L140: "… East Asia's NO_y concentrations during the winter-spring …" L479: "… primarily focused on NO_y loadings …" L507: "… provided seasonal snapshots of NO_y transport across East Asia …" L543: "… a comprehensive perspective on NO_y transport dynamics,

Comment	If I understood the manuscript right, the definition of NO_y used does not include NO_x. This makes sense if one would like to focus on transport, but it can be misleading if the results are interpreted in the way of "how much of the reactive nitrogen pollution is due to transnational transport". I think it would be better to include NO_x in NO_y. If that's not possible, the authors should at least include a column in the tables giving the fraction that transported NO_y contributes to $NO_y + NO_x$ assuming the latter is local.
Response	Thank you for pointing out this critical oversight. Our definition of NO_y includes NO_x, but we mistakenly did not state this. We have corrected this in the manuscript accordingly.
Changes made	L196-197: "Note that we used the summation of NO_x, nitric acid (HNO_3), nitrous acid (HONO), and peroxyacetyl nitrate (PAN) to represent NO_y …"

Comment	I'm completely confused by Figures 5 and 6. If I did not misunderstand the figures, they are supposed to show the absolute mixing ratio of NO_y that is present throughout East Asia and originates in one of the selected source regions. However, the maps show clear hotspots in polluted regions such as SMA even when the contribution is from another source region. Shouldn't these maps show smooth distributions outside the source regions? And shouldn't we see a reduction with distance from the source region? Please explain or correct the figures.
Response	We acknowledge that the discussions accompanying Figures 5 and 6 do not fully address the rationale behind the spatial distributions of NO_y, particularly the occurrence of hotspots outside the source regions. We'd like to clarify that these figures do not solely represent the physical transport of NO_y across the domain but rather illustrate where each source region's NO_x emissions contribute to local and transboundary NO_y concentrations. The formation of NO_y involves chemical reactions and transformations, which can lead to secondary hotspots outside the immediate vicinity of the source regions. To better understand these hotspots, we examined the spatial distributions of individual NO_y components. The earlier NO_y hotspots along the transport pathways seemed to correspond to longer-lived species such as HNO_3 and PAN, the formations of which depend on the availability of radicals and other precursors that are often more abundant in urban environments, which led to those hotspots pointed out. We have revised the manuscript to provide a more detailed explanation to support our discussions, accordingly.
Changes made	L433-441: "In addition, when using the a priori and a posteriori NO_x emissions during spring, the severity of NO_y pollution did not necessarily decrease as the distance from each source region increased… the formations of such species depend on the availability of radicals and other precursors that are often more abundant in urban environments, which emerged as these hotspots."

Comment	I do not follow the discussion on the impact of wind speed on the fraction of transported NO_y. In the manuscript, it is claimed that low wind speeds lead to accumulation of pollutants (so far, I agree) and that this leads to a larger contribution of transported NO_y. This I do not understand, as accumulation (and dilution) will work in a similar way for locally produced and transported NO_y, and therefore, the ratio is not changed. In my opinion, the only relevant quantities are a) how much NO_y is transported over the borders into the selected region and b) how much NO_y is produced within this region. Please explain. (From Minor comments) Lines 368 and 373: See my major comment on the impact of wind speed. I do not agree with the arguments made here about

	"rapid passing through" and "longer lingering" as wind speed affects all NO_y, not just the transported NO_y.
Response	Thank you for pointing this out. We acknowledge that the original discussion was self-contradictory in attributing a larger contribution of transported NO_y to weaker seasonal winds. Particularly, our statements were overdoing the relationship between the weakening Siberian High and slower exit of NO_y without supporting evidence. We recognize that accumulation due to weaker winds would affect both transported and locally produced NO_y similarly, and thus, the ratio between the two would not necessarily change as you commented. To address this, we have removed the unnecessary and misleading explanations in the discussion and revised the text to focus on the absolute quantities of NO_y transported across borders and produced locally within the selected regions. This clarification aligns with the reviewer's valid observation and ensures that the discussion reflects a more accurate interpretation of the results.
Changes made	L425: "… expanded westward noticeably …" L372-374 (in the original manuscript): "~~In addition, the highly stable and subsiding nature of the Siberian High could have trapped the NO_y in the lower boundary layer, further limiting the extent of their transport across longer distances (Zhang et al., 2007; Zhai et al., 2024).~~" L376-378 (in the original manuscript): "~~This can also be attributed to the slower exit of pollutants due to the less forceful winds, allowing pollutants to linger longer in receptor regions' stable atmospheres and increasing the transboundary impact (Wang et al., 2019; Lee et al., 2022).~~" "

Minor comments:

Comment	L149: While the PBL is an interesting region for air pollution studies, I'm surprised that the authors assume that it is also the altitude region where the relevant transport processes take place.
Response	Thanks for pointing this out and we agree that the original manuscript could mislead readers, as transport does not occur exclusively within the PBL. We have rephrased the relevant line and included additional clarifications in the methodology section.
Changes made	L191-195: "However, it is important to acknowledge that substantial pollutant transport also occurs in the free troposphere beyond the PBL, where stronger winds facilitate long-range movement of pollutants. Our study specifically focuses on the PBL to assess cross-regional pollutant behaviors, as this layer directly influences surface air quality, the modeled estimates of which can be evaluated with station measurements (detailed in Section 2.5), and human health associated."

Comment	L198: The sentence on the use of averaging kernels is rather vague and should be formulated more precisely.
Response	Thanks for pointing this out. We have elaborated further on how we made the direct comparison between the GEMS's and CMAQ's NO_2 columns in the manuscript.
Changes made	L248-264: "… including the averaging kernel, cloud fraction, data quality flags, … model-derived variables from the GEMS Level 2 data, including tropospheric and stratospheric air mass factors (AMF), a priori tropospheric NO2 profile, and tropospheric pressure profile from the WRF model coupled with Chemistry (WRF-Chem) 3.9.1 (NIER, 2020). … excluded the cloudy scenes which could lead to inaccurate AMF references".

Comment	L219: Note that GEMS does not take instantaneous snapshots of NO_2 but scans for 30 minutes from East to West.
Response	Thanks for your comment. We realized that our original description gives the nuance like GEMS's retrievals occur at such intervals. We have corrected the relevant statement in the manuscript accordingly.
Changes made	L285: "Due to the 15-minute offset in the availability of GEMS Level 2 products (from 22:45 to 07:45 UTC),"

Comment	L261: Introduce SMA
Response	We have added a description of the SMA earlier in the manuscript to provide some context before the discussion.
Changes made	L182-184: "During discussions for Korea (later in Section 3), we focused on the SMA, the country's economic hub, where dense traffic activity contribute to severe air pollution (Figure 1) (Park and Lee, 2020)."

Comment	Section 2.3: It is well known from validation studies that version 2 of the GEMS tropospheric NO_2 product as well as GEMS cloud fractions still have some issues. This should be briefly discussed and the impact on emissions and the quantification of NO_y transport be mentioned.
Response	Thanks for pointing it out, and we have added further explanation to support our use of the retrieval data that have a cloud fraction < 0.3, the criteria of which (as well as the problematic aspect in the dataset you have noted) was suggested in an earlier study that used GEMS Level 2 data version 2.0 (Lange et al., 2024).
Changes made	L262-264: "…note that since the Level 2 data version 2.0 quit employing the OMI climatology thereby deserves further validation efforts through retrieval studies, we excluded the cloudy scenes which could lead to inaccurate AMF references."

Comment	Figure 2: Are the model fields sampled at the times of valid GEMS measurements?
Response	Yes, we did not use any modeled scene outside the GEMS's retrieval times mentioned in the Methods to ensure their nearly fair comparisons against the retrieval data. We have clarified this aspect in the caption of the Figure 2.
Changes made	Figure 3 caption: "… Note that we excluded the modeled columns that do not correspond with the GEMS's retrieval times."

Comment	Figure 2: I do not understand why the model with a posteriori emissions overestimates NO_2 over large regions of Northern and Central China and Korea in February and March. With the assumption of local linearity, I would have expected the model to always do a good job on local hotspots such as SMA.
Response	Thanks for raising this important aspect regarding the overly adjusted a posteriori values. The modeled columns theoretically must become closer to the observed columns, but our employment of the Bayesian approach, which

	is often regarded as a simple inverse modeling method, could not fully resolve the transport effect (non-locality in the real world). We have added explanation for this aspect in the manuscript, accordingly.
Changes made	L345-348: “In addition, we noticed some posterior overcompensation in the modeled columns, shown by some overestimated values across North and South Central China and Korea … a simple inverse modeling method which cannot fully resolve the non-locality of air pollutants (Park et al., 2024) …”

Comment	Figure 2: What does the term “hourly” in the figure caption refer to?
Response	Thank you for pointing it out, and we realized that the caption is a little bit confusing. “Monthly average of hourly tropospheric NO_2 columns“ means the average of hourly tropospheric NO_2 columns for each month from January to May. We have revised the caption, accordingly.
Changes made	Figure 2 caption: “Averages of hourly tropospheric NO_2 columns (molecules/cm^2) observed and modeled during daylight hours (GEMS retrieval hours) in each month from January to May 2022”.

Comment	Line 323: A decline in anthropogenic emissions will increase the relative contribution of transported NO_y only if the other regions do not see a similar decline in emissions.
Response	Thank you for your comment. We realized that part of our earlier statement might have been misleading to readers. We have revised it accordingly in the manuscript.
Changes made	L403-405: “However, this does not fully explain the concurrent increase in transboundary contributions, suggesting that other factors, such as the weakening of meteorological barriers, facilitated broader dispersion of NO_y from source regions. These dynamics are discussed further below.”

Comment	Figure 4: Introduce ICO, BCO, OTH
Response	Thank you for pointing out the missing descriptions. We have introduced ICO, BCO, and OTH in the caption.
Changes made	Figure 4 caption: “ICO and BCO indicate the contributions from initial conditions and lateral boundary conditions, respectively, and OTH indicates the contribution of the emissions from the regions unspecified during ISAM”.

Comment	Figure 5: The ppb scale does not seem correct to me – 5x10E4 ppb of NO_y?
Response	Thanks for pointing it out, and we realized that our use of the ppb scale does not make sense when it comes to representing the summation of the pollutant loadings. We have corrected the unit from ppb to ton, which better explains such an aspect, accordingly.
Changes made	Figures 5, 6, and S2 units

Comment	Figure 5: What does the term total (sum) NO_y imply?
Response	The term indicates the total amount of NO_y accumulated during the winter months from January to February. We acknowledge that the term may confuse readers, so we have revised the captions.
Changes made	Figures 5 and 6 captions