Response to reviewers on: Numerical simulation of nitrous oxide over Asia using regional climate-chemistry-ecology coupling model RegCM-Chem-YIBs

Xin Zeng et al.

We sincerely thank the referees for their valuable comments, which have played a crucial role in improving the quality of this article. We have carefully revised and responded to them one by one. In this document, we describe how we have addressed the reviewers' comments. Review comments in black, responses in blue and text added/modified in manuscript in red.

RC1: 'Comment on egusphere-2025-608', Anonymous Referee #1

General comments:

Original comment 1#: Descriptions on how the climate and land models are lacking, particularly N_2O processes in the ecosystem model. My understanding is the integrated model has no nitrogen processes, as N_2O fluxes from CAMS and EDGAR are used as inputs, rather from internal simulations.

Response: We appreciate the reviewer's accurate understanding and helpful comment. Indeed, in the current study, the RegCM-Chem-YIBs framework does not include internal nitrogen cycle processes or dynamic N_2O production mechanisms within the land surface model. The N_2O fluxes are externally prescribed based on the EDGAR and CAMS inventories, without contributions from in-situ biogeochemical simulations (e.g., soil nitrification and denitrification N_2O fluxes). The primary objective of this study is to evaluate how different external inventories affect the simulated spatial and seasonal distribution of N_2O under a consistent atmospheric transport framework. We have now clarified this model limitation explicitly in the revised manuscript (Lines 143-160), and we have also emphasized this as a key direction for future model development in the Discussion section.

On the other hand, we have substantially revised the Introduction to improve its clarity, focus, and logical progression. We now begin by emphasizing the dual climate and ozone-depleting role of N_2O, establishing its relevance and urgency. We have removed general policy-related statements that were not directly aligned with the scientific focus of the paper. Through the literature, we found that a key gap in current modeling approaches: many frameworks treat the atmosphere as a passive boundary without explicitly simulating key atmospheric processes such as transport, mixing, and vertical redistribution of N_2O. Therefore, in order to assess the impact of surface emissions and atmospheric transport on the concentration of nitrous oxide (N_2O) in the troposphere, a three-dimensional global atmospheric chemical transport model (CTMs) is needed for research. We summarize recent advances in global-scale N_2O modeling and highlight that regional high-resolution simulations remain limited, especially over East Asia, a region with complex emission patterns and strong meteorological variability. Finally, we clearly stated the research objective: We used a chemically inert tropospheric tracer to separate and evaluate the impact of two inventories on N_2O concentration patterns in East Asia, driven by surface emissions and atmospheric transport, without considering internal biogeochemical processes of nitrous oxide. We also provided a detailed analysis of the seasonal variations and driving factors at six observation points in the region. These revisions improve the logical flow of the introduction and better position our study within the context of current scientific challenges. We believe the revised section more clearly conveys the rationale, novelty, and objectives of our work. We have now modified this part in the revised manuscript (Lines 34-126).

Revised version: (L143-149) In this study, we incorporate nitrous oxide (N_2O) as a new tracer species into the RegCM-Chem-YIBs modeling framework. This addition enables the simulation of N_2O concentration dynamics by considering external surface emissions and atmospheric transport processes. The model currently does not include stratospheric chemistry sinks of N_2O; hence, the simulated N_2O concentration is governed by its surface emissions and atmospheric redistribution. The surface N_2O emissions are prescribed from offline emission inventories (e.g., EDGAR and CAMS), which are read into the chemical module and mapped to model grids at each timestep. These inventories do not currently include the online biogeochemical nitrogen cycle from the land surface model (e.g., nitrification/denitrification in CLM4.5), but can be optionally replaced or supplemented by interactive fluxes in future versions. The atmospheric transport of N_2O follows the same numerical treatment as other long-lived tracers (e.g., CO_2, CH_4) in RegCM-Chem (Shalaby et al., 2012). The atmospheric transport includes advection and diffusion. Advection is based on the three-dimensional wind fields (u, v, w) provided by the RegCM dynamical core. The vertical diffusion is dominated by the eddy diffusivity coefficient computed from the planetary boundary layer (PBL) scheme. The eddy diffusivity coefficient depends on atmospheric stability, surface roughness, and PBL height, consistent with the scheme used for other trace gases. Horizontal diffusion is applied for numerical stability, using constant or latitude-dependent eddy diffusivities. Using the enhanced model, we simulate N_2O concentrations over East, South, and Southeast Asia in 2020 and analyze their seasonal and spatial patterns under different emission scenarios.

(Lines 34-126) Nitrous oxide (N_2O) ranks as the third most prevalent greenhouse gas in the atmosphere, following carbon dioxide (CO_2) and methane (CH_4). With a global warming potential 273 times that of CO_2 and an atmospheric lifetime of 116 ± 9 years (Prather et al., 2015), N_2O contributes approximately 6% to the radiative forcing from long-lived greenhouse gases (WMO, 2023; NOAA, 2024). By 2022, the global average concentration of N_2O had reached 335.8 ± 0.1 parts per billion (ppb), reflecting a 124% increase since the pre-industrial era. As a result, N_2O has become the largest contributor to the growth in effective radiative forcing among long-lived greenhouse gases since 1990 (WMO, 2023). In addition to its role as a potent greenhouse gas, N_2O is also a significant ozone-depleting substance (ODS). In the stratosphere, it undergoes photolysis to produce reactive nitrogen species (NOx), which catalyze ozone destruction (UNEP, 2013; Mcelroy and Mcconnell, 1971). In 2020, anthropogenic N_2O emissions, expressed in terms of CFC-11 equivalents, exceeded those of all CFCs by more than twofold, representing over 20% of the peak CFC emissions recorded in 1987 (Geneva, 2022). As such, N_2O is expected to remain the dominant ODS throughout the 21st century (Ravishankara et al., 2009). Despite its growing atmospheric burden, N_2O has received comparatively less attention than CO_2 and CH_4 in both observational and modeling communities. In particular, there is still a lack of clarity regarding its spatial and temporal distribution in the free troposphere and lower stratosphere, as well as the relative importance of emission patterns and atmospheric transport in shaping these distributions.

As a greenhouse gas of significant global concern, accurately simulating the emission and concentration distribution of N_2O is essential for assessing the impacts of climate change and crafting effective emission reduction strategies (De Sisto et al., 2024; Zhou et al., 2020). From the site-specific models such as DNDC (Li et al., 1992), DAYCENT (Parton et al., 1996) to the dynamic global vegetation models (DGVMs; Cramer et al., 2001) like ORCHIDEE (Krinner et al., 2005), O-CN (Zaehle and Friend, 2010), CLM (Saikawa et al., 2013), TRIPLEX-GHG (Zhang et al., 2017) , IBIS (Ma et al., 2022) and LPJ-GUESS (Ma et al., 2025), the detailed process of nitrous oxide generation is added to the global model, allowing N_2O emissions to be truly coupled to climate, carbon cycle, water cycle, and human activities, so that their source-sink changes and climate feedbacks can be assessed at a global scale. In addition to process-based modeling, other commonly employed approaches include the emission factor (EF) method recommended by the IPCC (IPCC, 2019), which estimates emissions from agricultural and industrial activities based on nitrogen input and empirically derived coefficients. This method remains widely used in national greenhouse gas inventories. Atmospheric inverse modeling represents another major technique, using top-down constraints from ground-based or satellite observations to optimize emission estimates (Fischer, 2015; Patra et al., 2022; Stell et al., 2022). Together, these bottom-up and top-down methods have greatly contributed to global N_2O budget assessments. However, most of these modeling frameworks treat the atmosphere as a boundary or diagnostic layer, lacking an explicit simulation of key atmospheric processes such as transport, mixing, and vertical accumulation. Given its long atmospheric lifetime (Prather et al., 2015), inter-hemispheric gradients, and sensitivity to large-scale circulation, explicitly modeling N_2O transport is crucial for accurate concentration assessments.

To assess the impact of surface emissions and atmospheric transport on tropospheric nitrous oxide (N_2O) concentrations, researchers have developed multiple three-dimensional global atmospheric chemical transport models (CTMs). Within the TransCom-N_2O project framework, models from multiple institutions differ in spatial resolution, vertical layering, meteorological forcing, and emission treatment, and have been widely employed to investigate the spatiotemporal distribution and transport mechanisms of N_2O (Thompson et al., 2014). The simulation based on GEOSCCM reconstructed the N_2O concentration and its isotope fluxes from 1980 to 2019, thereby revealing the contribution of anthropogenic emissions (Liang et al., 2022). Lickley et al. (2021) quantified the stratospheric influence on surface hemispheric differences in models and observations for N_2O. In addition, the influence of the Brewer-Dobson circulation on the transport and budget of N_2O in the stratosphere has also attracted attention (Minganti et al., 2022). Despite these advances, CTMs face persistent challenges. Most operate at relatively coarse spatial resolution (Chipperfield, 2006), limiting their ability to resolve regional and seasonal variations. Some models run in offline mode (Ishijima et al., 2010; Martin Heimann, 2003), relying on fixed meteorological inputs and thus unable to capture feedbacks between transport processes and emission changes. Furthermore, critical mechanisms such as stratospheric chemical reactions and stratosphere-troposphere exchange (STE) are often simplified or omitted. Therefore, enhancing the physical process characterization ability and spatial resolution of the model, as well as strengthening the coupling with observational data, are the key directions for future simulation of the evolution of N_2O concentration.

Regional climate-chemistry models, such as RegCM-Chem (Shalaby et al., 2012), provide the capability to resolve subcontinental-scale heterogeneity and capture dynamic meteorological drivers critical for simulating atmospheric constituents. However, their application to nitrous oxide (N_2O) remains limited and underexplored. While global inverse modeling studies have examined uncertainties associated with different inventories (Hong et al., 2017; Fischer et al., 2015), there is a notable lack of regional-scale evaluations on how discrepancies between inventories propagate through atmospheric transport to influence surface N_2O concentrations. This gap hampers our ability to accurately attribute observed concentration biases to either inventory errors or atmospheric process representations. Understanding the spatial and temporal variability of atmospheric N_2O at the regional scale is essential not only for assessing its climate forcing but also for its role in stratospheric ozone chemistry. East and South Asia, regions with substantial anthropogenic N_2O emissions driven by intensive fertilizer application and livestock production (De Sisto et al., 2024; Zhang et al., 2022), remain poorly studied in terms of how local emissions and atmospheric transport interplay to control observed N_2O variability. Observational and satellite data indicate that surface emissions, vertical mixing, and stratosphere-troposphere exchange collectively shape regional N_2O distributions (Nevison et al., 2011; Thompson et al., 2013). Yet, current high-resolution regional models often underrepresent these key processes, particularly in monsoon-influenced areas characterized by strong seasonal dynamics and vertical coupling. Therefore, advancing regional climate-chemistry modeling frameworks is critical to disentangle local versus transported signals, reduce uncertainty in regional N_2O budgets, and improve source attribution accuracy. Addressing these challenges will enable more robust evaluations of emission inventories and atmospheric processes, ultimately supporting more effective mitigation strategies for this potent greenhouse gas.

Here, we introduce N_2O as a transported inert tracer into the RegCM-Chem-YIBs regional climate-chemistry-ecosystem model. The implementation includes horizontal advection, vertical mixing, and convection of N_2O in the atmosphere, without internal biogeochemical sources or sinks. Two widely used inventories CAMS and EDGAR are used as surface forcing to drive the model. Our objectives are to assess the spatial and seasonal variability of simulated N_2O concentrations over East, South, and Southeast Asia in 2020, evaluate the sensitivity of model outputs to the choice of inventory and explore the driving factors influencing the seasonal concentration of nitrous oxide. Unlike many process-based studies focusing on terrestrial emissions, our approach decouples surface flux generation from atmospheric redistribution, thereby enabling a clearer assessment of how inventory choices affect modeled N_2O distributions. This work serves as an initial step toward the inclusion of N_2O in regional climate-chemistry modeling systems, with implications for understanding the atmospheric transport of N_2O in high-emission regions. It also provides a framework that can be extended in future studies to incorporate biogenic fluxes and feedbacks with land surface processes. Section 2 provides a detailed overview of the methods and datasets used in this study, while the subsequent results and discussions are analyzed in detail in Sections 3 and 4. Our main conclusions and summaries can be found in Section 5.

Original comment 2#: The manuscript focuses on the seasonal fluctuations of N_2O concentrations and concludes that surface concentration is low when surface N_2O emissions are low. This is not supported by their figures. Moreover, the seasonal variations across all sites are not "pronounced" as claimed by the authors. The fluctuations are quite small, within about 1~2 ppb, which is minor relative to the N_2O concentration. A deep analysis on seasonal fluctuations of atmospheric N_2O concentrations can refer to the paper "The Modeled Seasonal Cycles of Surface N_2O Fluxes and Atmospheric N_2O".

Response: Thank you for these important comments. We agree that our original figures did not clearly support the statement regarding the seasonal co-variation between N_2O emissions and concentrations. To address this, we conducted a quantitative correlation analysis between monthly mean surface N_2O concentrations simulated by the model and the corresponding surface N_2O emissions across the study domain. Figure S5 exhibits the correlation between the CAMS emissions and the simulated concentration is higher ($R^2 = 0.84$, $p < 0.01$), while the correlation for EDGAR is slightly lower ($R^2 = 0.46$, $p < 0.01$). We explain it in lines 456-470 of the revised edition.

We added a new section 2.5 named "Extraction of Seasonal Signals in Surface N_2O Concentration and Driving Factors". According to the method of Sun et al. (2024), this section elaborates in detail on how we detrend and standardize the observational data and model simulation results, as well as the specific methods for extracting the seasonal influencing factors of surface nitrous oxide concentration from the existing data. The new section was in the revised manuscript (Lines 246-291). Although the seasonal fluctuation range of nitrous oxide concentration is relatively small compared to its concentration, after detrending and standardizing the ground nitrous oxide concentration data, we found that the six stations within the study area showed a relatively obvious seasonality, with amplitudes reaching 2.9-3.6. Figure 8 shows the seasonal fluctuations in nitrous oxide concentrations across the six stations and the seasonal variations in driving factors. Table S3 lists the amplitudes of these factors at these sites, which can demonstrate their seasonal intensity and relative contributions. Importantly, our results are consistent with previous modeling studies. For instance, Nevison et al. (2007) showed that the modeled seasonal amplitude of surface N_2O generally represents only 0.1%–

0.2% of the annual mean value. Similarly, the N_2O seasonal amplitude of multiple sites selected by Sun et al. (2024) also rarely exceeds 2.5 ppb. In our study, the amplitude of seasonal cycles at individual stations (1–2 ppb) falls well within this expected range.

Revised version:

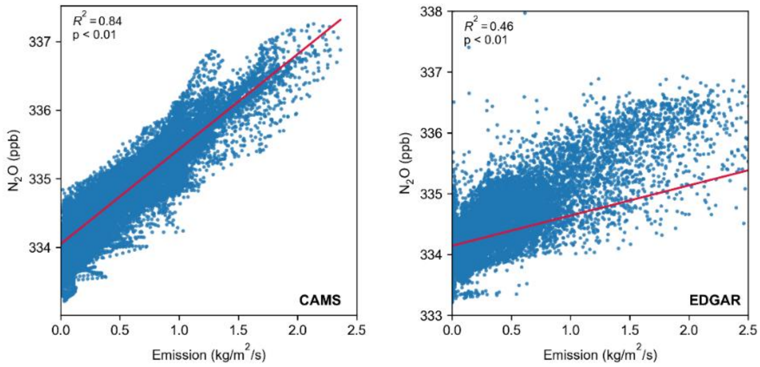

Figure S5. Spatial correlation of surface N_2O concentrations with emissions from CAMS and EDGAR inventories.

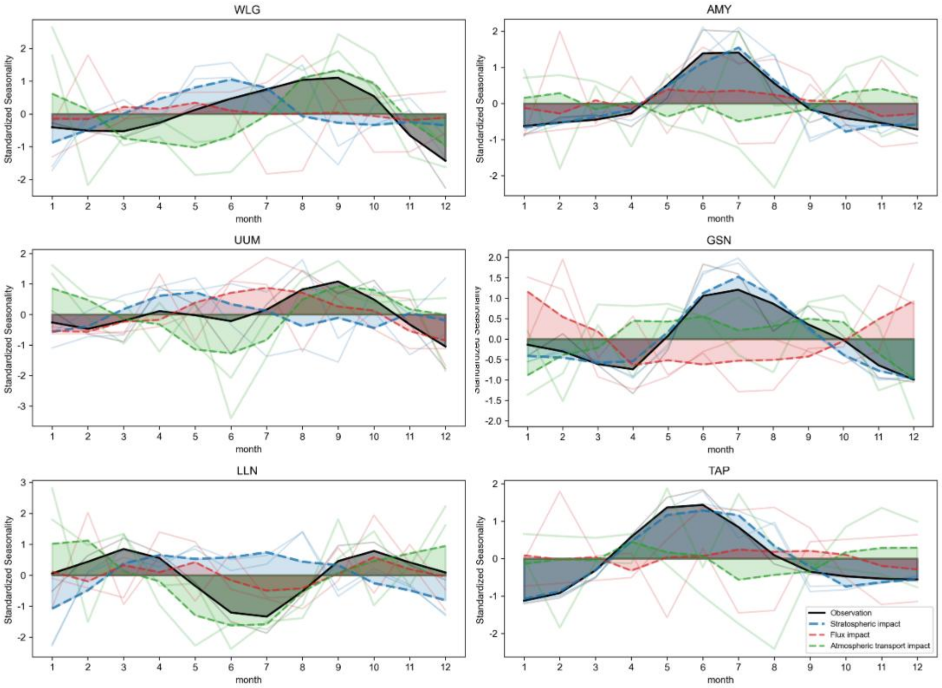

Figure 8. Seasonal patterns of surface N_2O concentrations and major driving factors at six sites in 2020.The black line represents the seasonal cycle of surface N_2O concentrations, while colored lines indicate the seasonal variations of influencing factors. Dashed red, blue, and green lines represent the smoothed average of the emissions contribution, stratospheric contribution, and atmospheric transport contribution from the two emissions results,

respectively. All variables are detrended and then normalized. For each factor, fine solid lines show the original data, and bold solid or dashed lines indicate the smoothed series. A shared legend is located in the lower-right corner of the TAP panel and applies to all six subplots.

Table S3. Amplitudes of seasonal cycles in surface N_2O concentrations and three major influencing processes: surface fluxes, atmospheric dynamics, and stratospheric contribution.

Site	observation	simulation	Flux impact	atmospheric transport impact	stratospheric impact
AMY	2.94	3.57	2.94	3.48	3.12
GSN	3.16	2.89	3.16	2.96	2.90
LLN	3.06	2.87	3.07	2.88	3.65
TAP	3.04	3.69	2.98	3.54	2.78
UUM	3.22	2.95	3.00	3.06	2.94
WLG	3.59	3.07	3.37	3.09	3.20

(L445-459): According to the grid-based correlation analysis results between the emission intensities from the two inventories (CAMS and EDGAR) and the simulated surface N_2O concentration field shown in Fig.S5, the results indicate that, within the study region, emission distribution is one of the primary drivers of the spatial variability in surface N_2O concentrations. The correlation between the CAMS emissions and the simulated concentration is higher ($R^2 = 0.84$, $p < 0.01$), while the correlation for EDGAR is slightly lower ($R^2 = 0.46$, $p < 0.01$). Despite the lower correlation, the EDGAR inventory remains valuable for large-scale assessments and policy-oriented studies, owing to its global consistency, regular data updates, and strong cross-regional applicability. It should be pointed out that although there is a strong spatial correlation, this does not imply a direct causal relationship between emission distributions and concentration patterns. Further comprehensive analyses incorporating transport processes and atmospheric dynamics are required. Importantly, the seasonality of surface N_2O concentrations exhibits pronounced spatial heterogeneity, particularly between land and ocean regions, where the dominant influencing factors differ significantly (Liao et al., 2004; Jiang et al., 2007; Sun et al., 2024). To better understand the region-specific drivers of this seasonality, we further investigated the seasonal patterns of N_2O at six ground-based observational sites across the study domain.

(L235-280): **2.5 Extraction of Seasonal Signals in Surface N_2O Concentration and Driving**

Factors

To investigate the seasonal variations of surface N_2O concentrations and their key driving factors, we performed detrending and standardization of the monthly N_2O time series (Sun et al., 2024) from six observational stations, as well as associated environmental drivers, including surface emissions, stratospheric influence, and atmospheric transport parameters.

Detrending

Detrending aims to remove long-term monotonic trends from each time series to isolate intra-annual variability. For a given monthly series x_t, detrending was done using least-squares linear regression, and the residual was defined as the seasonal anomaly:

$$x_t{}' = x_t - (\alpha t + \beta) \tag{1}$$

where α and β are the slope and intercept of the linear fit to x_t, and $x_t{}'$ is the detrended anomaly series. This procedure was applied to all relevant variables before seasonal comparison.

Standardization

To enable direct comparison of seasonal amplitudes and patterns among different variables which may have different units and variances, each detrended time series was further standardized using z-score normalization:

$$Z_t = (x_t{}' - \mu) / \sigma \tag{2}$$

where μ and σ are the mean and standard deviation of the detrended series $x_t{}'$. The resulting standardized time series Z_t has a mean of zero and unit variance, allowing direct comparison of seasonal fluctuations in N_2O and its drivers.

For the UUM site, observed surface N_2O concentrations were missing for November and December. These two values were gap-filled using linear interpolation to complete the seasonal cycle.

Decomposition of Seasonal Drivers

The seasonal variation in observed surface N_2O concentration ($\boldsymbol{S_{obs}}$) is known to be influenced by multiple processes (Nevison et al., 2011; Thompson et al., 2014), including:

- Surface emissions (EMI) from anthropogenic and natural sources
- Atmospheric transport (ATM) including advection and convection
- Stratospheric influence (STR) due to the stratosphere-troposphere exchange (STE) and chemical sinks in the stratosphere
- Model structural uncertainty and observation error ($\boldsymbol{\varepsilon}$)

We assume the seasonal signal of observed N_2O can be approximated as:

$$\boldsymbol{S_{obs} = S_{EMI} + S_{ATM} + S_{STR} + \varepsilon} \tag{3}$$

In our modeling system, RegCM-Chem-YIBs explicitly includes surface emissions and meteorology-driven transport, but stratospheric chemistry and STE are not. Thus, we approximate the stratospheric contribution (STR) to the seasonal cycle as the difference between observed and modeled N_2O at each site:

$$\boldsymbol{S_{STR} + \varepsilon \approx S_{obs} - S_{sim}} \tag{4}$$

where $\boldsymbol{S_{sim}}$ refers to the modeled N_2O concentration at the corresponding site. This residual implicitly includes the effects of STE, stratospheric sinks (e.g., photolysis, $O(^1D)$ reactions), and any model biases unrelated to emissions or transport and the observation error. For clarity, we refer to this residual as the stratospheric contribution (STC) in the following analysis.

The seasonality of surface emissions (EMI) is derived directly from the monthly gridded fluxes provided by the CAMS and EDGAR inventories. After regridding and matching to observation sites, the same detrending and standardization procedure was applied. The seasonal transport component (ATM) is then estimated by subtracting the flux-induced N_2O signal from the modeled total:

$$\boldsymbol{S_{ATM} = S_{sim} - S_{EMI}} \tag{5}$$

Original comment 3#: One advantage of regional climate models is that they can provide higher-resolution estimates of target variables. Yet there are no figures showing the model obtains N_2O concentrations with spatial details.

Response: We appreciate the reviewer's observation regarding the spatial resolution of the modeled N_2O concentration fields. Indeed, one of the key advantages of regional climate-chemistry models such as RegCM-Chem-YIBs is their ability to simulate atmospheric variables at fine spatial resolution (36 km × 36 km in our setup). However, in the original version, the figures based on simulations driven by CAMS reanalysis data (with a native resolution of 2.5° × 1.27°) appeared spatially coarse. This is because the input emissions and boundary conditions from CAMS, which are derived through global-scale top-down inversion, inherently limit the spatial variability of the resulting N_2O fields despite the high resolution of the dynamical core.

To better highlight the model's capacity to represent regional spatial detail, we have added new figures in the revised manuscript showing N_2O concentrations driven by the EDGAR inventory (resolution: 0.1° × 0.1°). These results clearly demonstrate enhanced spatial detail in surface N_2O distributions, especially over East Asia where emission heterogeneity is high. We believe this addition better supports the utility of the regional model and directly addresses the reviewer's concern. The revised figure is now shown in Figure 5.

Revised version:

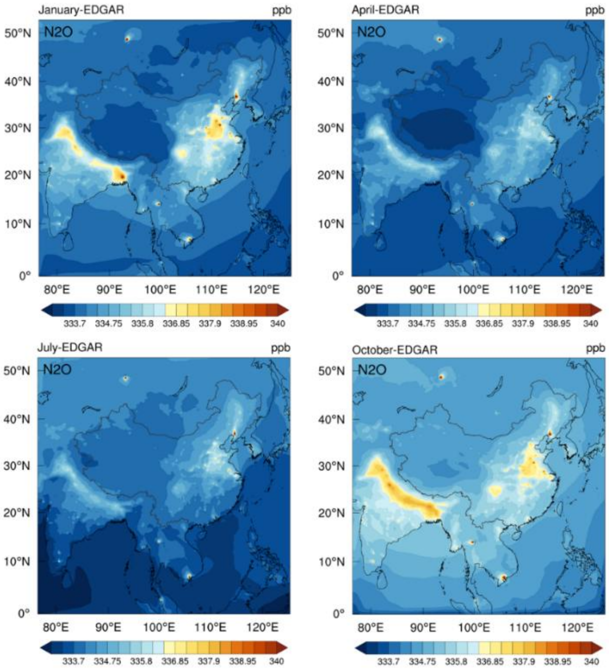

Figure 5. The distribution of the seasonal average ground-level N_2O concentration in 2020, as simulated by the EDGAR datasets.

Original comment 4#: The text is not well written. There are many statements explaining specific definitions or terms (e.g., Lines 109-112), which are not closely related to the topic of this manuscript.

Response: Thank you for comment. We fully agree that unnecessary definitions and explanations can detract from the clarity and focus of the manuscript. In the revised version, we have carefully reviewed the entire manuscript and removed or condensed background descriptions that are not directly relevant to the scientific objectives or methodology of this study. In addition, some necessary chapters and paragraphs have been added to broaden the depth of the article.

Specifically, we have revised Lines 109-112 to eliminate generic or widely known definitions. Our goal was to streamline the narrative and ensure that all included information directly supports the study's objectives and scope.

Additionally, we have improved the overall writing style for conciseness and scientific clarity.

Besides, we removed the overly general statements related to complex N_2O and atmosphere interactions and mitigation strategies, as these topics go beyond the scope of this study. We also revised the abstract and introduction to more accurately reflect the study objectives, emphasizing that this work serves as a necessary first step toward understanding the inventory driven uncertainties in N_2O simulations, prior to investigating interaction mechanisms or designing mitigation strategies. In addition, we explicitly clarified the study's scope in the introduction to avoid confusion about the intended contributions of the paper. We have made modifications to abstract in the revised manuscript (Lines 29-32, 118-124) to help readers better understand this work.

To study the seasonal variation of surface N_2O concentrations and the key drivers, we conducted detrending analysis and standardization on data of the six sites (Sun et al., 2024) and related environmental drivers (including surface emissions, stratospheric effects, and atmospheric transport parameters). We have added Section 2.5 to introduce the methods of detrending and standardization as well as the decomposition of factors affecting the seasonal signals of nitrous oxide on the ground. The modified parts are on lines 246-291 of the revised version.

We have divided the "results and discussions" in the original manuscript into two separate sections. For Section 3.4, we described in more detail the seasonality of nitrous oxide concentrations at the six sites in the study area and the contribution of its driving factors. After detrending and standardization, it was found that these sites all exhibited relatively obvious seasonal characteristics, and the contributions of each driving factor were not the same, varying from site to site. (Lines 456-511)

In the discussion section, we analyzed and compared the differences between the two inventories and the reasons for the similarities and differences between their simulation results and the observed and reanalyzed data. Then we discussed the reasons why the model of this study failed to reproduce the peak N_2O concentrations at the AMY, GSN and TAP sites in June. We also analyzed why the CAMS-driven simulations consistently produced higher N_2O concentrations than those driven by EDGAR at sites such as TAP, UUM, and WLG. In addition, the reasons why the regional average surface nitrous oxide concentration simulated by the model is low in summer and high in winter, which shows an inconsistent trend with the observed values of these six observation stations, were also discussed. We analyzed three possible reasons that could explain this phenomenon, which are seasonal variation in the height of the atmospheric boundary layer, regional differentiated transport effects and the STE influence. Finally, we discussed the uncertainties of the article, including the uncertainties of observations, model uncertainties, and the uncertainties of the decomposition methods of seasonal drivers, and analyzed the limitations of the model. In future work, we plan to incorporate more detailed internal biogeochemical processes of N_2O into the model framework, particularly soil nitrification and denitrification. Moreover, evaluating the influence of multiple emission inventories in parallel will enhance the robustness of our conclusions. To better constrain the vertical budget and assess the role of stratosphere–troposphere exchange, we also aim to adopt a fully coupled chemistry-climate model that explicitly resolves STE processes. These improvements will enable more accurate attribution of N_2O variability and support more reliable scenario projections. (Lines 521-649)

We hope these revisions help strengthen the manuscript's focus and improve its readability for readers.

RC2: 'Comment on egusphere-2025-608', Anonymous Referee #2, 03 Jul 2025

General comments:

Original comment 1#: The Introduction is not well developed and lacks a clear connection between the identified research gap and the specific research question addressed in this study. The motivation remains weak and unconvincing, and some background statements appear generic or only loosely related to the core topic of N_2O modeling (see specific comments below).

Response: We appreciate the reviewer's constructive feedback. We fully agree that the original introduction did not adequately articulate the link between the broader research gap and the specific scientific question addressed in this study. In response, we have substantially revised the Introduction to improve its clarity, focus, and logical progression.

We now begin by emphasizing the dual climate and ozone-depleting role of N_2O, establishing its relevance and urgency. We have removed general policy-related statements that were not directly aligned with the scientific focus of the paper. Through the literature, we found that a key gap in current modeling approaches: many frameworks treat the atmosphere as a passive boundary without explicitly simulating key atmospheric processes such as transport, mixing, and vertical redistribution of N_2O. Therefore, in order to assess the impact of surface emissions and atmospheric transport on the concentration of nitrous oxide (N_2O) in the troposphere, a three-dimensional global atmospheric chemical transport model (CTMs) is needed for research. We summarize recent advances in global-scale N_2O modeling and highlight that regional high-resolution simulations remain limited, especially over East Asia, a region with complex emission patterns and strong meteorological variability. Finally, we clearly stated the research objective: We used a chemically inert tropospheric tracer to separate and evaluate the impact of two inventories on N_2O concentration patterns in East Asia, driven by surface emissions and atmospheric transport, without considering internal biogeochemical processes of nitrous oxide. We also provided a detailed analysis of the seasonal variations and driving factors at six observation points in the region. These revisions improve the logical flow of the introduction and better position our study within the context of current scientific challenges. We believe the revised section more clearly conveys the rationale, novelty, and objectives of our work.

Original comment 2#: The current description of the N_2O-related processes in the model is insufficient and not acceptable in its present form for a model development and evaluation paper submitted to GMD. It lacks the necessary detail regarding the mathematical formulations, parameters, and model structure that have been added or modified. For instance, the statement "In this study, we introduce a new species of N_2O into the coupled model, taking into account the emissions, atmospheric transport, and diffusion processes" is far too vague. What exactly is "new"? How are N_2O emissions calculated? What are the mathematical representations of atmospheric transport and diffusion for N_2O in the model? How are these formulations different from those used for other gases, such as CO_2 or CH_4?

Response: We thank the reviewer for this insightful and constructive comment. We fully agree that the original description of the N_2O-related processes was too general and did not meet the expected level of technical detail for a model development paper. In response, we have substantially revised the manuscript to provide a more rigorous and complete description of the N_2O implementation in the RegCM-Chem-YIBs model.

We clarified that N_2O is introduced as a new passive tracer species into the chemical transport module of RegCM-Chem. This includes defining N_2O as a prognostic variable with its own transport equation, subject to emissions and atmospheric redistribution processes. We now explicitly state that N_2O emissions are derived from external offline inventories (e.g., EDGAR v8.0 and CAMS), and are provided to the chemical module at each model timestep. These emissions are currently not computed interactively within the land surface model, but this extension is planned for future work. The atmospheric transport includes advection and diffusion. Advection is based on the three-dimensional wind fields (u, v, w) provided by the RegCM dynamical core. The vertical diffusion is dominated by the eddy diffusivity coefficient computed from the planetary boundary layer (PBL) scheme. The eddy diffusivity coefficient depends on atmospheric stability, surface roughness, and PBL height, consistent with the scheme used for other trace gases. Horizontal diffusion is applied for numerical stability, using constant or latitude-dependent eddy diffusivities. We added text explicitly stating that the treatment of N_2O transport processes (advection, diffusion) is identical to those used for long-lived tracers like CO_2 and CH_4 in RegCM-Chem, thus ensuring internal consistency.

The revised description can now be found in Section 2.1: Model description (Lines 143-160), and we believe it now provides the level of technical clarity and completeness expected by GMD.

Original comment 3#: The authors have combined the Results and Discussion into a single section, which makes it difficult to distinguish between the presentation of model outputs and the interpretation or broader implications of other findings (e.g., Lines 224-232). This structure limits the clarity and depth of both components. I recommend splitting the Results and Discussion into separate sections. This would not only improve the organization of the manuscript but also provide space for a more focused and in-depth discussion of the results in the context of existing literature, model limitations, and uncertainties.

Response: Thank you for the insightful suggestion. In the original version, we combined the "Results" and "Discussion" sections to maintain a concise structure. However, we agree that this combined format may reduce clarity and analysis depth. In the revised original draft, we reorganized the content and split the original "Results and Discussion" section into two separate parts:

Section 3: Results. Now we focus on the presentation of model output, key results and comparative analysis. Section 3 is divided into four subsections, namely emission inventory differences and their impacts on modeled N_2O, model evaluation, spatiotemporal distribution of N_2O, and seasonality of ground N_2O concentrations.

Section 4: Discussion. We provided a more focused interpretation of the research results, evaluated the limitations of the model and experimental design, and delved into the reasons for the differences in the simulation effects of the two sets of lists. We analyzed the potential factors for the inconsistency between the site observations and the simulation results, as well as the uncertainties of this study, the shortcomings of the existing model, and the future development goals.

This reorganization enhances the readability of the manuscript and provides a clearer space for evaluating the robustness of our findings and placing them in a broader scientific context. We are grateful to the reviewers for their guidance, which has helped us enhance the clarity and organization of our manuscripts.

Original comment 4#: The main discussion section primarily highlights the agreement between the simulation results and previous studies, but it lacks a critical assessment of discrepancies or differences. A balanced discussion should also address where and why the model diverges from other findings, which is essential for understanding model behavior and performance in the future studies. Moreover, there is no discussion of model limitations. For a study published in a model development journal, it is important to transparently acknowledge the assumptions, uncertainties, and potential weaknesses in the model framework or input data.

Response: We thank the reviewer for this insightful comment. We agree that the original discussion lacked sufficient depth in analyzing discrepancies and explicitly acknowledging model limitations.

We have divided the “results and discussions” in the original manuscript into two separate sections. In the discussion section, we analyzed and compared the differences between the two inventories and the reasons for the similarities and differences between their simulation results and the observed and reanalyzed data. One reason is the limitation of model structure. The current model framework does not include the stratospheric photochemical loss processes of N_2O, which are implicitly accounted for in the CAMS reanalysis dataset through data assimilation. This omission leads to excessive accumulation of N_2O in regions influenced by stratosphere-troposphere exchange, causing an overestimation in CAMS-driven simulations. Another reason is the methodological differences in inventories. EDGAR is a bottom-up inventory based on activity data and emission factors, offering sectoral detail and emission-only signals but lacking temporal variability. CAMS, in contrast, is a top-down inversion constrained by observations. Its inversion process may conflate transport errors with emission estimates, introducing internal inconsistencies. Given our goal of isolating the role of emissions from transport, EDGAR provides a more suitable and interpretable signal for this modeling framework.

Then we discussed the reasons why the model of this study failed to reproduce the peak N_2O concentrations at the AMY, GSN and TAP sites in June. This deficiency is primarily attributed to the omission of key stratospheric processes, namely the chemical sink of N_2O and the stratosphere-troposphere exchange (STE). We also analyzed why the CAMS-driven simulations consistently produced higher N_2O concentrations than those driven by EDGAR at sites such as TAP, UUM, and WLG. We conducted a correlation analysis between the N_2O concentration simulated by the model and surface emissions. The results exhibited that within the study region, emission distribution is one of the primary drivers of the spatial variability in surface N_2O concentrations. The correlation between the CAMS emissions and the simulated concentration is higher ($R^2 = 0.84$, $p < 0.01$), while the correlation for EDGAR is slightly lower ($R^2 = 0.46$, $p < 0.01$). Compared with EDGAR, the emission intensity of these sites reported by the CAMS inventory is higher, which largely explains the reason for the increase in simulated concentrations.

In addition, the reasons why the regional average surface nitrous oxide concentration simulated by the model is low in summer and high in winter, which shows an inconsistent trend with the observed values of these six observation stations, were also discussed. We analyzed three possible reasons that could explain this phenomenon, which are seasonal variation in the height of the atmospheric boundary layer, regional differentiated transport effects and the STE influence. Finally, we discussed the uncertainties of the article, including the uncertainties of observations, model uncertainties, and the uncertainties of the decomposition methods of seasonal drivers, and analyzed the limitations of the model. In future work, we plan to incorporate more detailed internal biogeochemical processes of N_2O into the model framework, particularly soil nitrification and denitrification. Moreover, evaluating the influence of multiple emission inventories in parallel will enhance the robustness of our conclusions. To better constrain the vertical budget and assess the role of stratosphere-troposphere exchange, we also aim to adopt a fully coupled chemistry-climate model that explicitly resolves STE processes. These improvements will enable more accurate attribution of N_2O variability and support more reliable scenario projections.

Specific comments:

Original comment 1#: L18-22: Authors claim to gain the complex interactions between N_2O emissions and atmospheric processes, as well as to inform strategies for reducing N_2O emissions. However, these claims are not substantiated by the results presented. There is no discussion on the interaction mechanisms between N_2O dynamics and atmospheric processes, nor is there any explanation or evaluation of a developed strategy for N_2O mitigation.

Response: We sincerely thank the reviewer for the careful reading and constructive feedback. We agree that our original manuscript included overly broad claims that were not fully supported by the presented analyses. Our current work is primarily focused on evaluating how different N_2O inventories affect the simulated spatial and temporal distribution of near-surface

N_2O concentrations under a consistent atmospheric transport framework using the RegCM-Chem-YIBs model. To address this issue, we have made the following revisions: We removed the overly general statements related to complex N_2O and atmosphere interactions and mitigation strategies, as these topics go beyond the scope of this study. We also revised the abstract and introduction to more accurately reflect the study objectives, emphasizing that this work serves as a necessary first step toward understanding the inventory driven uncertainties in N_2O simulations, prior to investigating interaction mechanisms or designing mitigation strategies. In addition, we explicitly clarified the study's scope in the introduction to avoid confusion about the intended contributions of the paper. We have made modifications to abstract in the revised manuscript (Lines 29-32, 118-124) to help readers better understand this work.

Revised version:

Revisions to the abstract section (L29-32): These findings highlight the critical importance of transport dynamics and inventory uncertainties in shaping regional N_2O patterns. This study establishes a process-resolving framework for diagnosing atmospheric N_2O variability and sets the stage for future work incorporating coupled biogeochemical feedbacks.

Revisions to the introduction section (L118-124): Our objectives are to assess the spatial and seasonal variability of simulated N_2O concentrations over East, South, and Southeast Asia in 2020, evaluate the sensitivity of model outputs to the choice of inventory and explore the driving factors influencing the seasonal concentration of nitrous oxide. Unlike many process-based studies focusing on terrestrial emissions, our approach decouples surface flux generation from atmospheric redistribution, thereby enabling a clearer assessment of how inventory choices affect modeled N_2O distributions. This work serves as an initial step toward the inclusion of N_2O in regional climate-chemistry modeling systems, with implications for understanding the atmospheric transport of N_2O in high-emission regions. It also provides a framework that can be extended in future studies to incorporate biogenic fluxes and feedbacks with land surface processes.

Original comment 2#: L34-46: This section largely repeats the content of Lines 23-30 and reads more like a general background summary rather than a focused introduction to N_2O-specific modeling. Much of the text is common knowledge for the intended audience and does not add meaningful context to the study. I recommend the authors revise this part to be more concise and directly aligned with the scientific objectives of the paper. Avoid generic statements and instead focus on the specific research gap, methodological innovation, or model improvement being addressed. Redundant wording should be removed to improve clarity and relevance.

Response: Thank you for this valuable comment. We agree that the original paragraph contained overly general background information and did not clearly articulate the relevance to N_2O-specific modeling. In response, we have revised the first paragraph of the introduction to make it more concise and better aligned with the scientific focus of the study. The revised version (Lines 46-50) first briefly highlights the significance of N_2O and then transitions into the key knowledge gaps in its vertical and regional distributions, as well as the modeling challenges that motivate our work. We have also removed repetitive or generic statements to enhance clarity and relevance.

Revised version: Nitrous oxide (N_2O) ranks as the third most prevalent greenhouse gas in the atmosphere, following carbon dioxide (CO_2) and methane (CH_4). With a global warming potential 273 times that of CO_2 and an atmospheric lifetime of 116 ± 9 years (Prather et al., 2015), N_2O contributes approximately 6% to the radiative forcing from long-lived greenhouse gases (WMO, 2023; NOAA, 2024). By 2022, the global average concentration of N_2O had reached 335.8±0.1 parts per billion (ppb), reflecting a 124% increase since the pre-industrial era. As a result, N_2O has become the largest contributor to the growth in effective radiative forcing among long-lived greenhouse gases since 1990 (WMO, 2023). In addition to its role as a potent greenhouse gas, N_2O is also a significant ozone-depleting substance (ODS). In the stratosphere, it undergoes photolysis to produce reactive nitrogen species (NO_x), which catalyze ozone destruction (UNEP, 2013; Mcelroy and Mcconnell, 1971). In 2020, anthropogenic N_2O emissions, expressed in terms of CFC-11 equivalents, exceeded those of all CFCs by more than twofold, representing over 20% of the peak CFC emissions recorded in 1987 (Geneva, 2022).

As such, N_2O is expected to remain the dominant ODS throughout the 21st century (Ravishankara et al., 2009). Despite its growing atmospheric burden, N_2O has received comparatively less attention than CO_2 and CH_4 in both observational and modeling communities. In particular, there is still a lack of clarity regarding its spatial and temporal distribution in the free troposphere and lower stratosphere, as well as the relative importance of emission patterns and atmospheric transport in shaping these distributions.

Original comment 3#: L57: Stange et al. (2000) is not the original publication describing the N_2O development within the DNDC model. It would be great to refer to the original foundational work by Li et al. (1992; JGR), which first introduced the N_2O modeling framework in DNDC.

Response: Thank you for your valuable suggestion. We agree that Stange et al. (2000) is not the original reference for the development of the N_2O component in the DNDC model. Accordingly, we have replaced this citation with the foundational work by Li et al. (1992), which originally introduced the N_2O simulation framework in DNDC.

Revised version: Li, C., Frolking, S., and Frolking, T. A.: A model of nitrous oxide evolution from soil driven by rainfall events: 1. Model structure and sensitivity, Journal of Geophysical Research: Atmospheres, 97, 9759-9776, 10.1029/92jd00509, 1992.

Original comment 4#: L56-59: The citation of process-based N_2O modeling studies appears outdated. At a minimum, the authors should acknowledge the development and application of dynamic global vegetation models (DGVMs), such as O-CN, CLM, TRIPLEX, ORCHIDEE, LPJ-GUESS, CABLE, and DLEM, which have been widely used to estimate the global N_2O budget, as discussed in Tian et al. (2020). Furthermore, other key approaches, such as the IPCC-recommended emission factor (EF)-based method and atmospheric inversion modeling, are also essential tools for estimating N_2O emissions at regional and global scales. These approaches should be mentioned to provide a more comprehensive and up-to-date overview of current methodologies in the field.

Response: Thank you for this insightful comment. We agree that our initial review of process-based N_2O models was incomplete and did not adequately reflect recent developments in the field. In response, we have substantially revised this section of the manuscript (Lines 51–70) to provide a more comprehensive and up-to-date overview of current modeling approaches used to estimate N_2O emissions at regional and global scales. Specifically, we now highlight the growing role of dynamic global vegetation models (DGVMs) such as O-CN, CLM, TRIPLEX-GHG, IBIS, LPJ-GUESS, which integrate biogeochemical and climate feedbacks and are increasingly used for large-scale N_2O budget assessments. We also include mention of the IPCC-recommended emission factor (EF)-based approaches and atmospheric inversion methods, both of which have been widely adopted for independent constraints on N_2O source estimates.

To better structure this part, we organize the tools into four categories: (1) site-specific models like DNDC (Li et al., 1992) and DAYCENT (Parton et al., 1996), (2) dynamic global vegetation models (DGVMs; Cramer et al., 2001) like ORCHIDEE (Krinner et al., 2005), O-CN (Zaehle and Friend, 2010), CLM (Saikawa et al., 2013), TRIPLEX-GHG (Zhang et al., 2017) , IBIS (Ma et al., 2022) and LPJ-GUESS (Ma et al., 2025) (3) EF-based bottom-up approaches, and (4) top-down atmospheric inversion techniques. We further emphasize a key limitation shared by most of these models: they treat the atmosphere either diagnostically or as a boundary layer, lacking explicit simulation of transport, mixing, and vertical redistribution processes that are essential for understanding atmospheric N_2O variability. This limitation provides the motivation for our study, which integrates emission processes with atmospheric transport modeling in a unified framework.

Revised version: As a greenhouse gas of significant global concern, accurately simulating the emission and concentration distribution of N_2O is essential for assessing the impacts of climate change and crafting effective emission reduction strategies (De Sisto et al., 2024; Zhou et al., 2020). From the site-specific models such as DNDC (Li et al., 1992), DAYCENT (Parton et al., 1996) to the dynamic global vegetation models (DGVMs; Cramer et al., 2001) like ORCHIDEE (Krinner et al., 2005), O-CN (Zaehle and Friend, 2010), CLM (Saikawa et al., 2013), TRIPLEX-GHG (Zhang et al., 2017) , IBIS (Ma et al., 2022) and LPJ-GUESS (Ma et al., 2025), the detailed process of nitrous oxide generation is added to the global model, allowing N_2O emissions to be truly coupled to climate, carbon cycle, water cycle, and human activities, so that their source-sink changes and climate feedbacks can be assessed at a global scale. In addition to process-based modeling, other commonly employed approaches include the emission factor (EF) method recommended by the IPCC (IPCC, 2019), which estimates emissions from agricultural and industrial activities based on nitrogen input and empirically derived coefficients. This method remains widely used in national greenhouse gas inventories. Atmospheric inverse modeling represents another major technique, using top-down constraints from ground-based or satellite observations to optimize emission estimates (Fischer, 2015; Patra et al., 2022; Stell et al., 2022). Together, these bottom-up and top-down methods have greatly contributed to global N_2O budget assessments. However, most of these modeling frameworks treat the atmosphere as a boundary or diagnostic layer, lacking an explicit simulation of key atmospheric processes such as transport, mixing, and vertical accumulation. Given its long atmospheric lifetime (Prather et al., 2015), inter-hemispheric gradients, and sensitivity to large-scale circulation, explicitly modeling N_2O transport is crucial for accurate concentration assessments.

Original comment 5#: L78-85: The manuscript repeatedly emphasizes the importance of mitigating N_2O emissions and protecting the ozone layer. However, it is unclear whether this study directly addresses these broader issues in a meaningful or actionable way. If not, I recommend the authors avoid overusing such generic and broad statements. Instead, the focus should be on clearly articulating the specific research question being addressed and ensuring that the motivation aligns with the actual scope and objectives of the study. As it stands, the current presentation of the research motivation is not sufficiently compelling or focused.

Response: We thank the reviewer for this valuable feedback. We acknowledge that our original manuscript contained some broad statements emphasizing the importance of reducing N_2O emissions and protecting the ozone layer, which were not sufficiently linked to the specific scope and objectives of our study. In response, we have carefully revised the introduction and abstract sections to focus more precisely on the concrete research questions addressed by this work. Specifically, we now clearly articulate our main objectives: evaluating how different N_2O inventories influence simulated atmospheric concentrations within a consistent regional transport framework, and investigating the seasonal variations and driving factors of surface N_2O concentrations at selected observation sites in East Asia. By doing so, we have removed overly general claims and strengthened the research motivation to better reflect the actual contributions and limitations of our study.

Original comment 6#: L86-95: I believe it is necessary to include a paragraph summarizing the progress made by other regional climate-chemistry-ecosystem models (or similar modeling frameworks) regarding N_2O-related process development. This contextual background is important for readers to understand the advances achieved in previous studies and how the current work fits into or improves upon them. Additionally, is there a specific reason why 2020 was selected as the only study year?

Response: We appreciate the reviewer's insightful suggestion. We agree that providing such context is important for readers to understand prior advances and how our current work fits within or improves upon existing efforts. Accordingly, we have added a dedicated paragraph in the introduction that highlights key developments in three-dimensional global atmospheric chemical transport models (CTMs) which assess the impact of surface emissions and atmospheric transport on tropospheric nitrous oxide (N_2O) concentrations. The relevant revisions can be found in Lines 71-89.

Regarding the choice of the year 2020 for our simulation, we selected this year primarily because it is the most recent year with comprehensive and consistent emission inventories (CAMS, EDGAR) and available ground-based N_2O observations for model evaluation. Additionally, 2020 serves as a representative recent climatological year, enabling us to assess model performance under contemporary conditions.

Revised version: To assess the impact of surface emissions and atmospheric transport on tropospheric nitrous oxide (N_2O) concentrations, researchers have developed multiple three-dimensional global atmospheric chemical transport models (CTMs). Within the TransCom-N_2O

project framework, models from multiple institutions differ in spatial resolution, vertical layering, meteorological forcing, and emission treatment, and have been widely employed to investigate the spatiotemporal distribution and transport mechanisms of N_2O (Thompson et al., 2014). The simulation based on GEOSCCM reconstructed the N_2O concentration and its isotope fluxes from 1980 to 2019, thereby revealing the contribution of anthropogenic emissions (Liang et al., 2022). Lickley et al. (2021) quantified the stratospheric influence on surface hemispheric differences in models and observations for N_2O. In addition, the influence of the Brewer-Dobson circulation on the transport and budget of N_2O in the stratosphere has also attracted attention (Minganti et al., 2022). Despite these advances, CTMs face persistent challenges. Most operate at relatively coarse spatial resolution (Chipperfield, 2006), limiting their ability to resolve regional and seasonal variations. Some models run in offline mode (Ishijima et al., 2010; Martin Heimann, 2003), relying on fixed meteorological inputs and thus unable to capture feedbacks between transport processes and emission changes. Furthermore, critical mechanisms such as stratospheric chemical reactions and stratosphere-troposphere exchange (STE) are often simplified or omitted. Therefore, enhancing the physical process characterization ability and spatial resolution of the model, as well as strengthening the coupling with observational data, are the key directions for future simulation of the evolution of N_2O concentration.

Original comment 7#: L117-122: The ecological module (e.g., YIBs) should include a clear description of how other nitrogen-related dynamic processes, such as biological nitrogen fixation, wet/dry deposition, plant uptake, and hydrological N loss, are represented, especially in relation to N_2O production and loss. Without this level of process-level documentation, readers and future users cannot assess, reproduce, or compare the model implementation, which is a key requirement of GMD.

Response: We appreciate the reviewer's insightful comment. The current study employs the RegCM-Chem-YIBs model primarily due to its high spatial resolution and its capacity to simulate the interactions among air pollutants, greenhouse gases, and regional climate. In our implementation, we focus on the inclusion of nitrous oxide as a tropospheric inert greenhouse gas, similar to CO_2, which involves externally prescribed emissions and internally simulated atmospheric transport. The model does not contain the internal nitrous oxide source-sink process; hence, internal biogeochemical processes such as biological nitrogen fixation, plant uptake, and hydrological nitrogen loss are not represented. Instead, our study is designed to evaluate the impact of different prescribed N_2O inventories under the same atmospheric transport framework. Therefore, the scope of the study does not involve process-level simulation of N_2O production and loss but rather focuses on inventory-driven atmospheric concentration differences. We have clarified this point in the revised manuscript (Lines 143-160), to help readers better understand the model configuration and the intended focus of the study.

Revised version: In this study, we incorporate nitrous oxide (N_2O) as a new tracer species into the RegCM-Chem-YIBs modeling framework. This addition enables the simulation of N_2O concentration dynamics by considering external surface emissions and atmospheric transport processes. The model currently does not include stratospheric chemistry sinks of N_2O; hence, the simulated N_2O concentration is governed by its surface emissions and atmospheric redistribution. The surface N_2O emissions are prescribed from offline emission inventories (e.g., EDGAR and CAMS), which are read into the chemical module and mapped to model grids at each timestep. These inventories do not currently include the online biogeochemical nitrogen cycle from the land surface model (e.g., nitrification/denitrification in CLM4.5), but can be optionally replaced or supplemented by interactive fluxes in future versions. The atmospheric transport of N_2O follows the same numerical treatment as other long-lived tracers (e.g., CO_2, CH_4) in RegCM-Chem (Shalaby et al., 2012). The atmospheric transport includes advection and diffusion. Advection is based on the three-dimensional wind fields (u, v, w) provided by the RegCM dynamical core. The vertical diffusion is dominated by the eddy diffusivity coefficient computed from the planetary boundary layer (PBL) scheme. The eddy diffusivity coefficient depends on atmospheric stability, surface roughness, and PBL height, consistent with the scheme used for other trace gases. Horizontal diffusion is applied for numerical stability, using constant or latitude-dependent eddy diffusivities. Using the enhanced model, we simulate N_2O concentrations over East, South, and Southeast Asia in 2020 and analyze their seasonal and spatial patterns under different emission scenarios.

Original comment 8#: L124-134: In the Experimental Design section, it is unclear whether any sensitivity analyses were performed to identify which parameters or input drivers (or climate variables) have the greatest influence on the simulated N_2O concentrations at different pressure levels, particularly in comparison with site-level observations. If such tests were conducted, please specify which sensitivity analysis method was used (e.g., one-at-a-time, Monte Carlo, Sobol, etc.) and summarize the key findings. Additionally, it would be better to clarify how the model was spun up prior to the start of the 2020 experiment or protocol runs. Details on the spin-up duration, initialization datasets, and criteria for equilibrium should be provided.

Response: We acknowledge the reviewer's suggestion regarding sensitivity analyses. While we did not conduct a formal sensitivity analysis (e.g., one-at-a-time or global variance-based methods), our study includes a comparison of two widely used N_2O inventories (EDGAR and CAMS), which can be viewed as a scenario-based experiment to assess how differences in emission inputs influence modeled N_2O concentrations. However, we agree that this does not constitute a systematic sensitivity analysis in the strict methodological sense, and we have revised the manuscript to clarify this distinction. We have added this clarification in the revised manuscript (Lines 170-185).

Due to the lack of internal sources and sinks of N_2O in the model, a full multi-year spin-up was not conducted. Because N_2O has a long atmospheric lifetime and is regarded as chemically inert in this setting, the model was initialized using CAMS to reanalyze the N_2O concentration dataset. Short-term initialization is sufficient to capture the concentration response to the specified surface flux and atmospheric transport. The simulation was initialized using chemical and meteorological fields from the boundary conditions provided by The CAMS global inversion-optimized greenhouse gas fluxes and concentrations dataset and ERA-Interim reanalysis dataset, respectively, starting from January 1st, 2020. To sum up, given that the N_2O is not chemically reactive in the troposphere and that no internal source/sink processes are included in this setup, the influence of spin-up is expected to be limited, especially for concentration comparisons at seasonal scales. We have added this clarification in the revised manuscript (Lines 165-169).

Revised version: The simulation period spans the full calendar year of 2020 and no spin-up was applied due to the lack of internal sources and sinks of N_2O in the model. Because N_2O has a long atmospheric lifetime and is treated as chemically inert in this setup, the model was initialized with CAMS reanalysis N_2O concentration dataset. The short-term initialization is sufficient to capture concentration responses to prescribed surface fluxes and atmospheric transport.

Two parallel simulation experiments were conducted, differing only in the N_2O inventory used as surface forcing:

CAMS Case: Using the CAMS inversion-optimized N_2O fluxes.

EDGAR Case: Using the bottom-up EDGAR anthropogenic N_2O emissions.

The physical and chemical processes in the model are configured using a combination of widely validated parameterization schemes. Gas-phase chemistry is represented by the CBM-Z mechanism (Zaveri and Peters, 1999), while planetary boundary layer processes are modeled using the Holtslag scheme (Holtslag et al., 1990). Convection is simulated with the Grell cumulus parameterization (Grell, 1993), and land-atmosphere interactions are resolved using the CLM4.5 land surface model (Oleson et al., 2008; Stöckli et al., 2008). Radiative processes follow the CCM3 radiation scheme (Zhang et al., 1998; Giorgi et al., 2012). These configurations are consistent with prior studies employing RegCM-Chem-YIBs in East Asia (Xie et al., 2019; Ma et al., 2023). All other boundary conditions, physical parameterizations, and model configurations were held constant between the two runs, ensuring that differences in simulated N_2O distributions resulted solely to differences in the inventories. Inner natural N_2O sources from soils and oceans were excluded, enabling a targeted investigation of input anthropogenic signals.

Original comment 9#: L203-207: I believe this part belongs to "Methodology" section.

Response: We agree with you. We remove this part from the section3.1.

Besides, we added a new section2.5 named "Extraction of Seasonal Signals in Surface N_2O Concentration and Driving Factors". This section elaborates in detail on how we de-trend and standardize the observational data and model simulation results, as well as the specific methods for extracting the seasonal influencing factors of surface nitrous oxide concentration from the existing data. The new section was in the revised manuscript (Lines 246-291).

Revised version:

2.5 Extraction of Seasonal Signals in Surface N_2O Concentration and Driving Factors

To investigate the seasonal variations of surface N_2O concentrations and their key driving factors, we performed detrending and standardization of the monthly N_2O time series (Sun et al., 2024) from six observational stations, as well as associated environmental drivers, including surface emissions, stratospheric influence, and atmospheric transport parameters.

Detrending

Detrending aims to remove long-term monotonic trends from each time series to isolate intra-annual variability. For a given monthly series x_t, detrending was done using least-squares linear regression, and the residual was defined as the seasonal anomaly:

$$x_t{}' = x_t - (\alpha t + \beta) \tag{1}$$

where α and β are the slope and intercept of the linear fit to x_t, and $x_t{}'$ is the detrended anomaly series. This procedure was applied to all relevant variables before seasonal comparison.

Standardization

To enable direct comparison of seasonal amplitudes and patterns among different variables which may have different units and variances, each detrended time series was further standardized using z-score normalization:

$$Z_t = (x_t{}' - \mu) / \sigma \tag{2}$$

where μ and σ are the mean and standard deviation of the detrended series $x_t{}'$. The resulting standardized time series Z_t has a mean of zero and unit variance, allowing direct comparison of seasonal fluctuations in N_2O and its drivers.

For the UUM site, observed surface N_2O concentrations were missing for November and December. These two values were gap-filled using linear interpolation to complete the seasonal cycle.

Decomposition of Seasonal Drivers

The seasonal variation in observed surface N_2O concentration ($\boldsymbol{S_{obs}}$) is known to be influenced by multiple processes (Nevison et al., 2011; Thompson et al., 2014), including:

- Surface emissions (EMI) from anthropogenic and natural sources
- Atmospheric transport (ATM) including advection and convection
- Stratospheric influence (STR) due to the stratosphere-troposphere exchange (STE) and chemical sinks in the stratosphere
- Model structural uncertainty and observation error ($\boldsymbol{\varepsilon}$)

We assume the seasonal signal of observed N_2O can be approximated as:

$$\boldsymbol{S_{obs} = S_{EMI} + S_{ATM} + S_{STR} + \varepsilon} \tag{3}$$

In our modeling system, RegCM-Chem-YIBs explicitly includes surface emissions and meteorology-driven transport, but stratospheric chemistry and STE are not. Thus, we approximate the stratospheric contribution (STR) to the seasonal cycle as the difference between observed and modeled N_2O at each site:

$$\boldsymbol{S_{STR} + \varepsilon \approx S_{obs} - S_{sim}} \tag{4}$$

where $\boldsymbol{S_{sim}}$ refers to the modeled N_2O concentration at the corresponding site. This residual implicitly includes the effects of STE, stratospheric sinks (e.g., photolysis, $O(^1D)$ reactions), and any model biases unrelated to emissions or transport and the observation error. For clarity, we refer to this residual as the stratospheric contribution (STC) in the following analysis.

The seasonality of surface emissions (EMI) is derived directly from the monthly gridded fluxes provided by the CAMS and EDGAR inventories. After regridding and matching to observation sites, the same detrending and standardization procedure was applied. The seasonal transport component (ATM) is then estimated by subtracting the flux-induced N_2O signal from the modeled total:

$$S_{ATM} = S_{sim} - S_{EMI} \tag{5}$$

Original comment 10#: L243-262: The evaluation of temperature, humidity, and wind in the model reads somewhat abrupt, as it appears without prior explanation or context. It is unclear why these three variables are evaluated alongside N_2O concentrations. I guess these are key climatic drivers influencing the simulated N_2O at different pressure levels? this should be clearly justified upfront. Ideally, the authors should first demonstrate the strength of their influence on N_2O through sensitivity analyses (as previously suggested), and then present the evaluation results accordingly.

Response: Thank you for this helpful comment. We agree that the evaluation of meteorological variables (temperature, humidity, and wind) was not well integrated into the structure of Section 3.2 and lacked a clear connection to the main focus of N_2O simulation. Therefore, we have revised this section by removing the evaluation of these meteorological fields and focusing on the model's performance in simulating N_2O concentrations. Specifically, we now emphasize comparisons with surface observations from ground-based sites and CAMS N_2O reanalysis data. Our original motivation for evaluating the meteorological variables was to ensure the reliability of the background meteorological fields, which underpin the atmospheric transport of N_2O. Although this information is valuable, we agree that it is better presented elsewhere or used as supporting context when discussing the physical drivers of N_2O transport, rather than being included in the model evaluation section. We have accordingly revised the manuscript to improve the clarity and focus of Section 3.2. Changes have been made in Lines 373-386 of the revised manuscript.

Revised version: In addition to site-based observations, the simulated N_2O field was also compared with the CAMS global reanalysis concentrations for January and July 2020 (Fig.4). Both datasets show consistent spatial gradients, with higher N_2O concentrations in densely populated areas such as eastern China and northern India. In major emission hotspots, the concentrations simulated by EDGAR are typically 1-5 ppb higher than those of CAMS. The July results of the two inventories showed a very high degree of consistency. However, in the January simulation, compared to EDGAR-driven results, CAMS slightly overestimates concentrations in northwest China but underestimates concentrations in high-emission areas near eastern China and the Ganges River Basin in India. As shown in Fig. S2, during spring and summer when surface N_2O concentrations are low, the model performs very well, with no obvious overestimation or underestimation. However, during autumn and winter, when the concentrations of nitrous oxide are relatively high, the results of CAMS input data overestimates high-emission areas in autumn and underestimates them in winter. The spatial distributions of annual high-emission areas in autumn and winter obtained from EDGAR input data are largely consistent but underestimated compared to the reanalysis data.

Original comment 11#: L270-281: The presentation of the site-level comparison results is overly descriptive and lacks in-depth analysis. I recommend the authors focus more on the discrepancies in seasonality between simulations and observations, which would be more interesting for readers. For instance, why does the model fail to reproduce the N_2O peak in June at sites such as AMY, GSN, and TAP? Additionally, what causes the simulated N_2O concentrations driven by CAMS to be consistently higher than those driven by EDGAR at TAP, UUM, and WLG?

Response: We appreciate the reviewer’s suggestion and agree that more comprehensive analysis of the seasonal discrepancies between simulations and observations would improve the manuscript. Accordingly, we have revised the structure of the manuscript by splitting the original Section 3 “Results and Discussion” into two separate sections: section 3 “Results” and section 4 “Discussion”.

In the “Discussion” section, we explored in detail the possible reasons why the model failed to capture the June N_2O peak at AMY, GSN, and TAP. This deficiency is primarily attributed to the omission of key stratospheric processes, namely the chemical sink of N_2O and the stratosphere-troposphere exchange (STE).

We also discussed why the CAMS-driven simulations consistently produced higher N_2O concentrations than those driven by EDGAR at sites such as TAP, UUM, and WLG. We conducted a correlation analysis between the N_2O concentration simulated by the model and surface emissions. The results exhibited that within the study region, emission distribution is one of the primary drivers of the spatial variability in surface N_2O concentrations. The correlation between the CAMS emissions and the simulated concentration is higher ($R^2 = 0.84$, $p < 0.01$), while the correlation for EDGAR is slightly lower ($R^2 = 0.46$, $p < 0.01$). Compared with EDGAR, the emission intensity of these sites reported by the CAMS inventory is higher, which largely explains the reason for the increase in simulated concentrations.

In addition, the reasons why the regional average surface nitrous oxide concentration simulated by the model is low in summer and high in winter, which shows an inconsistent trend with the observed values of these six observation stations, were also discussed. We analyzed three possible reasons that could explain this phenomenon, which are seasonal variation in the height of the atmospheric boundary layer, regional differentiated transport effects and the STE influence.

These modifications have significantly improved the clarity and depth of the analysis. This part is in lines 542-576 of the revised version.

Revised version: The model in this study failed to reproduce the peak N_2O concentration at the AMY, GSN and TAP sites in June. This deficiency is primarily attributed to the omission of key stratospheric processes, namely the chemical sink of N_2O and the stratosphere-troposphere exchange (STE). Previous studies have shown that STE plays a critical role in shaping the seasonal variability of surface N_2O (Thompson et al., 2011). These sites are located within the East Asian monsoon influence area, where strong summer monsoon circulation and enhanced convection promote the downward transport of N_2O-depleted stratospheric air, resulting in the surface N_2O concentration reaching a significant trough in September (Ruiz and Prather, 2022).

The absence of this STE mechanism in the model leads to a dampening or complete loss of seasonal peaks, underscoring the importance of representing cross-tropopause transport in regional simulations. In addition, we observe that CAMS-driven simulations consistently yield higher surface N_2O concentrations than those driven by EDGAR at TAP, UUM, and WLG. According to our correlation analysis between observed N_2O concentrations and surface emissions, surface flux remains an important driver of variability (Tian et al., 2020). As shown in Fig. 2b, the CAMS inventory reports higher emission intensities at these sites compared to EDGAR, which largely explains the elevated simulated concentrations. These findings are in line with previous work demonstrating that uncertainties in emission inventories can significantly influence modeled N_2O concentrations (Saikawa et al., 2014; Thompson et al., 2019).

It is worth noting that although individual sites such as AMY, GSN, and TAP exhibit a summer-high and winter-low concentration pattern, the regional average over East Asia shows the opposite trend that lower N_2O concentrations in summer and higher in winter. This apparent contradiction can be explained by the following three mechanisms: Seasonal variation in the height of the atmospheric boundary layer: In summer, the height of the boundary layer in East Asia deepens considerably, diluting near-surface N_2O and lowering the regional average concentration. In contrast, a shallow winter boundary layer facilitates pollutant accumulation, resulting in higher concentrations (Jaffe et al., 1999). Regional differentiated transport effects: Background or high-altitude sites are less affected by direct anthropogenic emissions. In contrast, the regional average includes more urban and lowland areas where N_2O accumulation is enhanced in winter due to stagnant meteorological conditions and regional transport (Thompson et al., 2014). The STE process of nitrous oxide is enhanced during convection-active summers, and stratospheric air transport to the surface may dilute surface N_2O concentrations but have a certain hysteresis effect, and low values of nitrous oxide may be observed at the surface in late summer (Ruiz and Prather, 2022). Since the stratospheric nitrous oxide sink and STE processes are not added to the model, the model does not reflect this stratospheric contribution, resulting in different patterns. However, the actual effect at specific sites depends on the strength and vertical extent of mixing (Nevison et al., 2004).

Original comment 12#: L309-310 & L336-338: This sentence is repetitive and does not add new information, consider removing it.

Response: Thank you for comment. We removed L336-338 from the original version.

All the revisions have been incorporated into the revised manuscript, and we hope that the updated version now meets the requirements of the journal. We sincerely appreciate the reviewer's thoughtful comments and efforts, and we respectfully invite a re-evaluation of the manuscript.

The following are the newly added references:

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