Anonymous Referee #2

- ***Snow is a key component of the cryosphere and has significant impacts on surface energy balance, hydrology, atmospheric circulation, and more. Moreover, snow is significant in atmospheric chemistry, where snow impurities such as nitrate are sensitive to sunlight and can be photolyzed to emit reactive species including NO2 and HONO, which can significantly disturb atmospheric chemistry, especially in pristine regions. An accurate description of the emission and atmospheric consequences of snow-emitted reactive species is hence important for assessing the atmosphere environment. To address this issue, the authors parameterized atmospheric nitrate deposition and its distributions in snow using WRF-Chem model, the performance of the simulations in snow depth, and BC, dust and nitrate concentrations are well validated by field observations in northern China. Overall, this paper is well written and will be very helpful to the related research communities to improve the understanding of snow-atmosphere interactions and its influence on environments. I think this work is suitable for publication in GMD if the following concerns can be addressed:***

Response: We are grateful to the reviewer for his/her time and efforts reviewing this manuscript. We have carefully read through the comments and made responses as well as relevant revisions in the manuscript. Please find our point-by-point response (black) and the corresponding revisions (blue) below.

Major comments

- ***1. You point out that to simulate snow nitrate photolysis and its impacts on overlying atmospheric chemistry, one need to obtain snow cover, snow depth, and snow physical and chemical properties, including snow density; impurities, including BC, dust; and nitrate. Other studies have parameterized most factors except snow nitrate concentration, which was the primary contribution of your work. However, your title was "Simulations of Snow Physicochemical Properties in Northern China using WRF-C", and the abstract includes much descriptions about the simulation and validations of snow cover, snow depth, and BC and dust concentrations, which ware not belonging to your work. In contrast, the description about snow nitrate simulation, the primary contribution of your work, was not enough. So, I suggest some necessary revisions to the title and abstract to emphasis your highlight on snow nitrate simulation. For example, in your abstract, the quantitative performance in nitrate concentration simulation, the bias analysis, and possible bias sources should be included to show the readers how good is your simulation. In addition, the results should more focus on snow nitrate simulation.***

Response: We sincerely thank the reviewer for the insightful comments and suggestions. We agree with the reviewer that the title and abstract should more clearly emphasize the primary contribution of our work, specifically the snow nitrate simulation. In light of this, we have revised the title and abstract to reflect this focus.

The new title is:

"WRF-Chem simulations of snow nitrate and other Physicochemical Properties in Northern China

"

The new abstract is:

"Snow is a key component of the cryosphere and has significant impacts on surface energy balance, hydrology, atmospheric circulation, and etc. In addition, numerous studies have indicated that snow impurities, especially nitrate, are sensitive to sunlight and can be photolyzed to emit reactive species including NO_2 and HONO, which serve as precursors of O_3 and radicals and disturb the overlying atmospheric chemistry. This makes snow an important reservoir of reactive species, especially in remote and pristine regions with limited anthropogenic emissions. The magnitude of snow chemical emissions is also influenced by snow physical properties, including snow depth, density and concentrations of light-absorbing impurities (e.g., BC and dust). Exploring and elucidating the emissions and atmospheric consequences of the snow-sourced reactive species require a global or regional model with a snow module. Here, we parameterized atmospheric nitrate deposition and its distributions in snow using a regional chemical transport model, i.e., the WRF-Chem (Weather Research and Forecasting Model coupled with Chemistry) model, and evaluated its performance in simulating snow cover, snow depth, and BC, dust, and nitrate concentrations with field observations in northern China, one of the regions with dense and prolonged snow cover. In general, the model simulated spatial variability of nitrate mass concentrations in the top snow layer (hereafter NITS) are consistent with observations. Simulated NITS values in Northeast China from December 2017 to March 2018 had a maximum range of 7.11–16.58 $\mu g\ g^{-1}$, minimum range of 0.06–0.21 $\mu g\ g^{-1}$, and a four-month average of 2.72 ± 1.34 $\mu g\ g^{-1}$. In comparison, observed values showed a maximum range of 9.35–33.43 $\mu g\ g^{-1}$, minimum range of 0.09–0.51 $\mu g\ g^{-1}$, and an average of 3.74 ± 5.42 $\mu g\ g^{-1}$. The model results show an underestimation especially in regions closes to large cities in northeastern China, most likely due to the underestimation of NO_x emissions in these regions. Additionally, nitrate deposition, snowpack accumulation processes, and challenges in capturing fine-scale emission variability may also contribute to the bias. The results illustrate the ability of WRF-Chem in simulating snow properties including concentrations of reservoir species in northern China, and in the future, we will incorporate snow nitrate photolysis in the model, exploring the emissions of snow NO_x from nitrate photolysis and the impacts on local to regional atmospheric chemistry and air pollutant transformations."

The descriptions on method were not clear:

- ***(a) Line 180, from Equations 2 and 3, horizontal diffusion was not concluded in wet deposition calculation, is its influence was insignificant?***

Response: Thanks for the comment. In WRF-Chem, horizontal diffusion of chemical species is included during transport processes. By the time wet deposition is calculated, chemical concentrations have already been influenced by horizontal diffusion, so its effects are inherently included in the deposition calculation.

- ***(b) Line 192, form Equation 4, the unit of MNITS should be same to $\Delta F \times dtime/\Delta Wsno$. However, in Equation 5, the unit of MNITS was same to $\Delta F \times dtime/\Delta Wsno \times \Delta t$.***

Response: Thanks for your careful check. We are sorry to make this mistake. Here we multiplied by an extra Δt. The term Δt already represents a cumulative value for the wet and dry deposition which means that the additional multiplication by Δt was incorrect. We have revised the formula by removing it in the Equation 5 (now Equation 7) as: "

$$M_{NITS}^{new} = M_{NITS} + \frac{\Delta F \times dtime}{\Delta W_{sno}} \quad (7)$$"

- ***(c) Line 193, ΔF is the cumulative wet and dry deposition of atmospheric nitrate during the entire period between the newly fallen snow and the previous time step. This means the unit of ΔF was kg m-2. If so, the unit of the second term in Equation 5 was not kg kg-1. Please check.***

Response: Thank you for pointing out the discrepancy. We have corrected it.

- ***(d) Line 219-220, you mentioned "the nitrate concentrations in each snow layer are determined by factors such as atmospheric deposition rates, the amount of new snowfall, layer combinations and divisions, and meltwater flushing (Oleson et al., 2010b; Flanner et al., 2012; Flanner et al., 200 ", how did you consider the layer combinations and divisions in your simulation.***

Response: We thank the reviewer for the comments. In our simulation, which calculates nitrate concentrations in snow, when new snowfall occurs, if a snow layer has nearly melted or its thickness falls below the minimum threshold, it is combined with the adjacent upper or lower layer to streamline the simulation. Conversely, if the snow layer exceeds the maximum thickness, it is subdivided into two layers of equal thickness, retaining the liquid water, ice content, and temperature of the original layer. This approach follows the snow layering system from SNICAR, which is based on the thermal layers used for thermodynamic calculations in the CLM land surface model (Flanner et al., 2012; Flanner and Zender, 2005; Flanner et al., 2007; Oleson et al., 2010). The model typically defines snow layer thicknesses as follows: the surface layer spans 0–3 cm, the second layer 3–7 cm, the third layer 7–18 cm, the fourth layer 18–41 cm, and the bottom layer exceeds 41 cm.

- ***(e) Line 185, you assumed a mix with the top 2cm layer? Do you have any references? For dry deposition, I can agree with your assumption, but for wet deposition, such an assumption may induce significant bias.***

Response: Thank you for pointing this out. We apologize for the lack of clarity in our original text. We intended to express that, upon deposition, nitrate is mixed instantly and uniformly in the model surface layer, which never exceeds 3 cm in thickness. In the original manuscript, we mistakenly stated 2 cm. We have now revised the sentence in the revised manuscript:
Page 7, Line 185 in the original manuscript: "After deposition, nitrate is mixed instantly and uniformly in the model surface layer, which never exceeds 3 cm thick." This indicates that we are adding nitrate to the model's surface layer, which is defined as 0-3 cm based on the SNICAR layering approach (Flanner et al., 2012; Flanner and Zender, 2005; Flanner et al., 2007; Oleson et al., 2010).

- ***(f) Line 212, the scavenging ratio for nitrate was assigned to 0.2. From my knowledge, the nitrate was much soluble. Assigning a low scavenging ratio should add more***

discussions.

Response: We agree with the reviewer that nitrate is highly hydrophilic and easily soluble in water. The value of 0.20 used in our study is derived from previous assumptions made by Flanner et al. (2012) and Zhao et al. (2014) regarding BC, which are generally reasonable compared to observations (Doherty et al., 2013). We acknowledge that this assumption may be oversimplified, as the value for nitrate is uncertain, and 0.20 may indeed underestimate the scavenging of nitrate. However, for this process to be effectively impactful, significant melting would need to occur. During our simulation period, temperatures in northern China were consistently low, primarily below 0 ° C, and significant melting did not take place. Therefore, we believe the impact of this assumption is minimal in this context. We have now included a more detailed discussion in the revised manuscript, specifically in Section 2.2.2:

Page 8, Line 211 in the original manuscript: " *D* represents the combined effect of total atmospheric particulate and gaseous nitrate deposition, which is specifically added to the surface layer of the snowpack. In this study, following Flanner et al. (2012) and Zhao et al. (2014), the scavenging ratio (k) for nitrate is assumed to be 0.2. This value is highly uncertain for nitrate and needs to be constrained by future observations (Flanner et al., 2012; Qian et al., 2014; Zhao et al., 2014). However, for this process to be effectively impactful, significant melting would need to occur. During our simulation period, temperatures in northern China were consistently low, primarily below 0°C, and significant melting did not take place. Therefore, we believe the impact of this assumption is minimal in this context. It is worth noting that the portion of nitrate mass lost through meltwater from the bottom layer of snow is considered to be removed from the snowpack and is not accounted for within the model."

- ***(g) In Equation 6, what did qi+1ci+1 represent, please clarify.***

Response: Thanks for this comment. In Equation 6 (now Equation 9), qi+1ci+1 represents the mass flux of water leaving layer *i*+1 (the layer above) multiplied by the concentration of nitrate in that same layer *i+1*. This term accounts for the transfer of nitrate from the upper layer to the current layer *i*, as meltwater moves downward through the snowpack. Similarly, qici represents the mass flux of water leaving layer i multiplied by the nitrate concentration in layer i. The change in nitrate mass in layer i influenced by the amount of nitrate coming from layer i+1 and the amount leaving layer *i*. We have clarified this in the revised manuscript as "The term $q_{i+1}c_{i+1}$ represents the mass flux of water leaving the layer above (i+1) multiplied by the concentration of nitrate in that layer, accounting for the transfer of nitrate from the upper layer to the current layer."

Minor comments:

- ***Suggest to add scatter plots of simulated versus observed data for simulation validations, especially for snow nitrate.***

Response: Thanks for this suggestion. Another reviewer had a similar comment, and we have now added scatter plots for snow surface nitrate in the revised manuscript as shown in Fig. 11c.

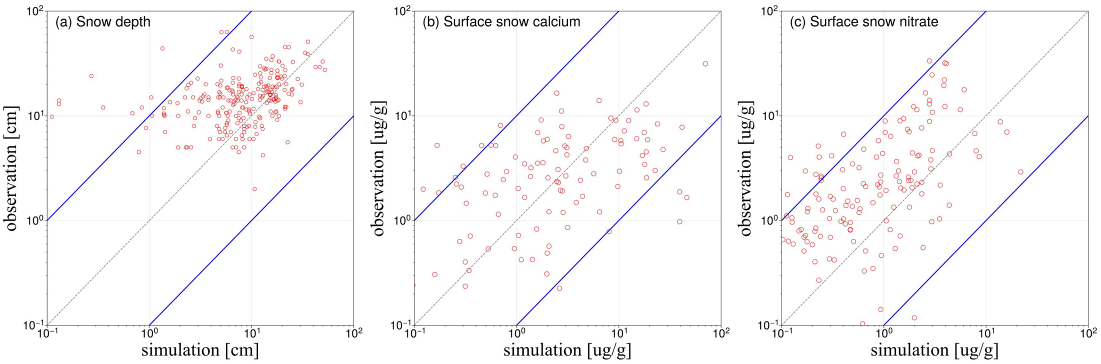

Figure 11. Scatter plots of the observations of **(a)** snow depth (cm), **(b)** surface snow calcium ion concentrations （ug/g） and **(c)** surface snow nitrate concentrations (ug/g) versus the corresponding WRF-Chem simulations in winter 2017–2018.

However, from Fig. 11c, we notice that the model generally underestimates the surface snow nitrate concentrations. Therefore, we further discussed the possible reasons for this underestimation. We have added this discussion to Section 3.4.2 in the revised manuscript as following:

From Page 22, Line 505 in the original manuscript:

"Regarding this underestimation, as illustrated in Figure 9, we note that there is a low bias for the NITS in high-pollution areas between December 2017 and January 2018. In particular, in high-pollution regions like Jilin Province, the model exhibited a negative bias, with an average observation-to-simulation ratio of 1.7, corresponding to a Normalized Mean Bias (NMB) of 40.29%. ~~This discrepancy might be attributed to the accumulation processes within the snowpack. However, it is important to acknowledge that the model also encompasses various uncertainties, such as incomplete representations of emission sources, nitrate production mechanisms, deposition processes, and aerosol scavenging in snow.~~

In addition, we extracted the simulated values corresponding to the observations at each station and plotted them as a scatter plot (Fig. 11c). The results show that the model generally underestimates the NITS. Typically, such an underestimation of NITS could result from either underestimating the amount of snow or underestimating the flux of nitrate deposition within the snow. However, based on the snow depth simulation results, the snow amount simulation performs better, so snowfall is unlikely to be the main cause of this bias. The most likely reason for this underestimation may be that the modeled atmospheric nitrate concentration is lower than the actual concentration. Consequently, even with the same snowfall amounts, the nitrate deposition would be underestimated. To demonstrate this, we analyzed the observed atmospheric nitrate concentrations from Tracking Air Pollution in China (Geng et al., 2017; Liu et al., 2022) and compared them with the simulated results. We found that in northern China, where our study area is located, the simulated atmospheric particulate nitrate concentrations were indeed lower than the observed values (Fig. S3). The low simulated nitrate concentrations in northern China may be due to incomplete atmospheric nitrate chemistry in the model. However, in other regions of southern China, such as Anhui (29.45° N - 34.55° N, 114.95° E - 119.55° E) and Fujian (23.65° N - 28.25° N, 115.95° E - 120.45 ° E), the simulated atmospheric nitrate concentrations closely matched the observations (Fig. S4). Thus, the effect of incomplete atmospheric nitrate chemistry in the model can be excluded in this case. Another possible reason for the low simulated nitrate concentrations in northern China could be the underestimation of NO_x emissions in this region. We also compared the observed and modeled atmospheric NO_2 concentrations in this region and found that the model indeed underestimated the NO_2 concentrations (see Fig. S5). In conclusion, the underestimation of NITS in the model is most likely due to the underestimation of atmospheric nitrate concentrations, which probably originates from the model's underestimate of NO_x emissions in this region.

In addition to analyzing the top snow layer, we further evaluated the model's performance by comparing the vertical distribution..."

- ***Line 415, BCS increase during the melting period should be mainly due to melt enrichment (Doherty et al., 2013)***

Response: Thank you for this comment. We agree that the rise in BCS should primarily be attributed to melt enrichment, as referenced by Doherty et al. (2013). We have now revised the text to clarify this point as "As the snow begins to melt, BCS continues to rise primarily due to melt enrichment, where melting snow concentrates BC near the snow surface (Doherty et al., 2013). This effect is further enhanced by dry deposition until the snow completely melts."

- ***Line 506-509 add necessary references to support your discussions.***

Response: Thank the reviewer for this suggestion. Based on the previous comments and the inclusion of the scatter plot, we have accordingly revised the discussion, and the original content from Lines 506-509 has been removed. However, the relevant points have been addressed in the conclusion, where we have added the necessary references related to the content from the original Lines 506-509.

- ***Suggest to add more quantitative bias analysis, especially for Section 3.4.2 Nitrate concentrations and spatial distribution.***

Response: We thank the reviewer for the suggestion. Another reviewer had a similar comment. Now we have added specific quantitative values to the bias analysis in Section 3.4.2 as "In particular, in high-pollution regions like Jilin Province, the model exhibited a negative bias, with an average observation-to-simulation ratio of 1.7, corresponding to a Normalized Mean Bias (NMB) of 40.29%."

- ***Line 543-549 add necessary references to support your discussions.***

Response: We thank the reviewer for this suggestion. We have incorporated necessary references and, based on previous comments and the inclusion of the scatter plot, we have accordingly revised the discussion. The new statement of this part is as follows:

From Page 23, Line 543 in the original manuscript: "The most likely reason for the discrepancies in NITS between the model and observations is the underestimation of atmospheric nitrate concentrations, which probably originates from the model's underestimate of NO_x emissions in this region. Additionally, uncertainties in the deposition processes (Akter et al., 2023; Huang et al., 2015; Lu and Tian, 2014), including dry and wet deposition of nitrate from the atmosphere to the snowpack, could also play a role. Furthermore, post-depositional processes could further contribute to the differences between the model and observations. These processes include snowfall dynamics, snow accumulation, and gas and aerosol scavenging in the snow (An et al., 2022; Flanner et al., 2012; Li et al., 2022; Poschlod and Daloz, 2024; Qian et al., 2014; Zhao et al., 2014), all of which may introduce uncertainties in the simulation of NITS. Another factor contributing to these discrepancies could be the relatively coarse model resolution, as it may not sufficiently capture the heterogeneous spatial distributions of snow and nitrate concentrations, especially when fine-scale variations are significant (Berg et al., 2024; Yu, 2013)."

- ***Line 516-519 you mentioned comparing simulated monthly values with observed daily values should be cautioned due to the significant temporal fluctuations in NITS, why did you do daily-to-daily comparisons as you can output daily results.***

Response: Thanks for your comment. Here we mentioned it is not a comparison of model monthly values versus observed daily values. Although snow samples are collected on specific dates, they do not represent only the conditions of that day. Snow accumulation is a cumulative process, and the data collected reflects the conditions accumulated over a certain period prior to the sampling date. The challenges mentioned here arise from the difficulty in determining whether an observation accurately represents conditions from the past few days, weeks, or even a month. Indeed, while the model can output daily results, the observed data does not represent daily conditions. Instead, the samples reflect an accumulation of conditions over the past few days or weeks.

- ***Line 543-549 is repeated by Line 555-563 more or less, please simplify.***

Response: We sincerely thank the reviewer for careful review and valuable suggestions. We have now deleted and merged the redundant content in the conclusion section. Also, we have incorporated necessary references and, based on previous comments and the inclusion of the scatter plot, we have accordingly revised the discussion. The new statement of this part is as follows:

From Page 23, Line 543 in the original manuscript: "The most likely reason for the discrepancies in NITS between the model and observations is the underestimation of atmospheric nitrate concentrations, which probably originates from the model's underestimate of NO_x emissions in this region. Additionally, uncertainties in the deposition processes (Akter et al., 2023; Huang et al., 2015; Lu and Tian, 2014), including dry and wet deposition of nitrate from the atmosphere to the snowpack, could also play a role. Furthermore, post-depositional processes could further contribute to the differences between the model and observations. These processes include snowfall dynamics, snow accumulation, and gas and aerosol scavenging in the snow (An et al., 2022; Flanner et al., 2012; Li et al., 2022; Poschlod and Daloz, 2024; Qian et al., 2014; Zhao et al., 2014), all of which may introduce uncertainties in the simulation of NITS. Another factor contributing to these discrepancies could be the relatively coarse model resolution, as it may not sufficiently capture the heterogeneous spatial distributions of snow and nitrate concentrations, especially when fine-scale variations are significant (Berg et al., 2024; Yu, 2013). Overall, however, the model demonstrates its ability in capturing the temporal and spatial variations in snow impurity concentrations including nitrate in Northern China. The considerable daily and diurnal fluctuations in simulated NITS emphasize the need for caution when comparing average values derived from the model with observations, as practiced in certain global modeling analyses.

(Huang et al., 2011; Qian et al., 2014; Zhao et al., 2014). It's worth mentioning that despite the overall evaluation of simulated spatial patterns of snow depth and aerosol concentrations within the snowpack against observations, discrepancies persist in simulating snow impurities, even though they have been fully quantified. Such biases could stem from uncertainties across various snow model processes, the primary emission sources of impurity precursors from atmosphere model, the gas and aerosol scavenging in the snow, and etc. To ensure accurate representation of aerosol contents within the snow, it is essential for the model to effectively simulate the life cycle of aerosols within snowpack, as highlighted in previous studies by Flanner et al. (2012) and Qian et al. (2014). Furthermore, uncertainties in the SNICAR model parameters must be quantified and constrained through observational data. Additionally, it is crucial for the model to precisely replicate the atmospheric aerosol life cycle, encompassing the faithful representation of atmospheric aerosol levels and the accurate treatment of deposition mechanisms. Improvements in such model parameters and mechanisms would be necessary to further improve the agreement with observations. Moreover, other factors such as atmospheric chemistry mechanisms may also need to be improved to better represent nitrate chemistry, which will be addressed in the next phase of this study.

Given the reasonable agreements between the model and observations, we will further incorporate snow nitrate photolysis and the subsequent emissions of NO2 and/or HONO to the overlying atmosphere, investigating the potential disturbs on local to regional atmospheric chemistry with focuses on aerosol burden which is important for atmospheric and snow radiative balances in snow cover regions, and on the potential effects on air quality originating from the winter snow cover to the downwind regions in Northern China.”

Reference

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