We would like to thank the reviewer for the constructive comments really helping to improve the paper. Below, we address each comment in full detail. Following the Reviewers' comments in black, please find our point-to-point responses in blue. Hereafter, all new added or modified sentences are marked in blue and italic in this response. The line numbers in the responses to comments are in accordance with the revised manuscript.

Reviewer #1

Xie et al. applied the WRF and CAMx models over Northeast China to investigate how chlorine chemistry in the atmosphere and on the snow-covered ground influenced atmospheric concentrations of N_2O_5, $ClNO_2$, $PM_{2.5}$ and O_3. They modeled February and March of 2024 when atmospheric measurements are available for N_2O_5 and $ClNO_2$ at a suburban location in Changchun City, Northeast China. They modified CAMx to compare two heterogeneous chemistry schemes (YU20 and BT09) that parameterize reactive uptake of N_2O_5 and HCl (from coal burning emissions) to aerosol surfaces. They also utilize the CAMx surface chemistry model to investigate how reactive uptake of N_2O_5 and HCl by snowpack may influence atmospheric N_2O_5 and $ClNO_2$. Xie et al. find that the YU20 scheme performed better than BT09 at describing the observed concentrations of N_2O_5 and $ClNO_2$. They find that reactive uptake of N_2O_5 by aerosol (YU20 scheme) is more influential than reactive uptake to snowpack and adding chlorine/chloride emissions (ACEIC inventory) in combination. They note that $ClNO_2$ formation from N_2O_5 uptake is influential on model results for PM2.5, ozone and other oxidants. They suggest that future modeling studies (for similar winter conditions) should include these emissions and processes. This study expands on previous 1D modeling studies by presenting a 3D picture of model sensitivity to algorithms and input data. The results can provide useful guidance for modeling similar conditions and planning future field campaigns.

Q1: section 2.5 (and elsewhere) I understood "anthropogenic chlorine emissions" to mean specifically the ACEIC inventory, especially gaseous HCl from coal combustion. Does the MEIC emission inventory include emissions of particulate chloride (PCl)? The ISORROPIA scheme in CAMx equilibrates HCl and PCl depending on aerosol pH and consequently emissions of both PCL and HCl contribute to available reactive chloride. Most likely, simulation Y1 includes some anthropogenic chloride emissions (i.e., PCl from MEIC) and the other simulations have more chloride/chlorine emissions (i.e., from ACEIC). The manuscript should clarify whether MEIC includes chloride emissions. Add a table summarizing mass of chloride/chlorine emissions from MEIC and ACEIC. Provide a citation for ACEIC when first mentioned.

Response:

Thank you for the insightful comments. In response to your questions, we provide the following clarifications:

1. Does the MEIC inventory include emissions of particulate chloride (PCl)?

The MEIC anthropogenic emission inventory does include PCl as a chlorine-containing species. However, in our emission preprocessing, we multiplied the emission factor of PCl from MEIC by zero, effectively setting its emissions to zero. This was done to avoid double-counting with PCl emissions from the ACEIC inventory. Consequently, in all simulations (including Y1), MEIC contributes no anthropogenic chlorine emissions (neither PCl nor HCl, etc.). All anthropogenic chlorine emissions used in the simulations are solely from the ACEIC inventory.

2. Summary table of chlorine emissions from MEIC and ACEIC

The table below (Table S1) summarizes the total emissions of chlorine-containing species from the ACEIC and MEIC inventories. As shown, all chlorine species from MEIC are either zero or not present, while ACEIC includes emissions of HCl, PCl, Cl_2, and HOCl.

Table S1 Chlorine emissions from the ACEIC and MEIC anthropogenic inventories used in this study (unit: Gg)

Inventory	HCl	PCl	Cl2	HOCl
ACEIC	361.34	173.72	17.55	79.45
MEIC	—	0.00	—	—

(Note: MEIC only contains PCl among chlorine species, but its emissions are set to zero; other species are not included, denoted by "—".)

3. Citation for the ACEIC inventory

A reference for the anthropogenic chlorine emissions inventory (ACEIC) has been added at the first mention in the manuscript (line 155).

Q2: The CAMx surface model can store pollutant mass within the snowpack, and this mass can be lost to the ground, e.g., via meltwater. Was loss of chloride from the snowpack in meltwater modeled, and was it influential?

Response:

Thank you for raising this question regarding the loss of chloride from snowpack via meltwater and its potential influence. Our response is as follows:

In the CAMx surface model, the original framework includes three surface types: soil, vegetation, and snow. After dry and wet deposition, atmospheric species deposited to the surface can undergo the following fates: (1) adsorption onto various media (e.g., vegetation, snowpack, and soil); (2) heterogeneous chemical reactions and photolysis, with some products subsequently re-emitted back to the atmosphere; and (3) loss through other processes, such as snowmelt (denoted as SnoMlt) (Figure1). The surface module allows users to specify the species and reaction pathways of interest (see schematic in Figure 2). In this study, we included N_2O_5, HCl, Cl, HNO_3, $ClNO_2$ and NO_2 in this module to represent the relevant surface reactions (see R1-R2).

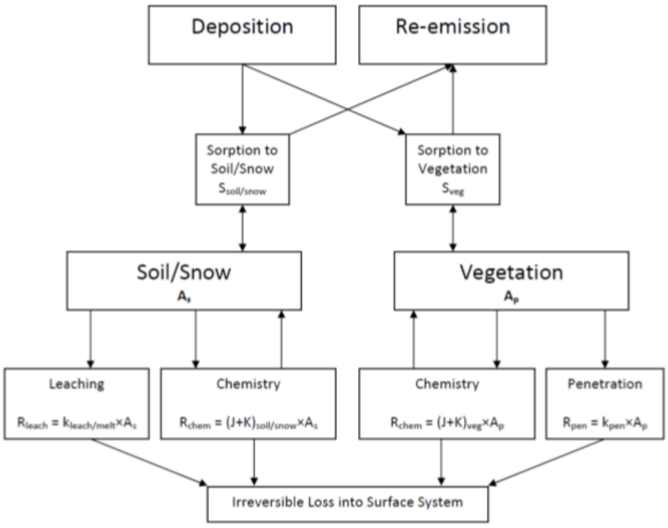

Figure 1. Schematic of the CAMx surface model (Ramboll., 2020).

$$HCl + N_2O_5 \xrightarrow{Ground} ClNO_2 + HNO_3 \quad \text{(R1)}$$

$$ClNO_2 + hv \xrightarrow{Ground} Cl + NO_2 \quad \text{(R2)}$$

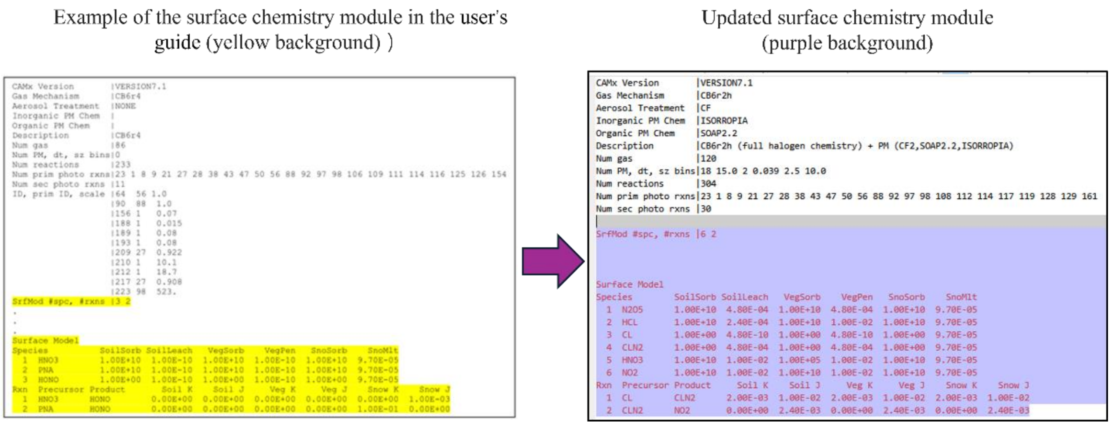

Figure 2. Configuration of the CAMx surface model.

Regarding the snowmelt process, the model incorporates two snowpack parameters (snow age and snow depth) to capture the dynamic evolution of snow accumulation and ablation (Figure 3). It is important to note that WRF-CAMx is a three-dimensional atmospheric chemical transport model that focuses primarily on the dynamic changes of pollutants in the atmosphere. The surface module is designed such that deposited species, after chemical transformation, are re-emitted to the atmosphere; the model only calculates the fluxes of these re-emitted species into the atmospheric column. Consequently, while the model does simulate the chemical transformation and re-release of chloride species during snowmelt, it does not account for the mass lost downward via meltwater to the soil or groundwater—such loss falls outside the gas-phase mass balance framework of the model and therefore does not directly influence the atmospheric chemical analysis in this study.

We have added the above explanation and the relevant schematic figures to help readers clearly understand the model's treatment of snowpack processes and its inherent limitations. We appreciate your careful review.

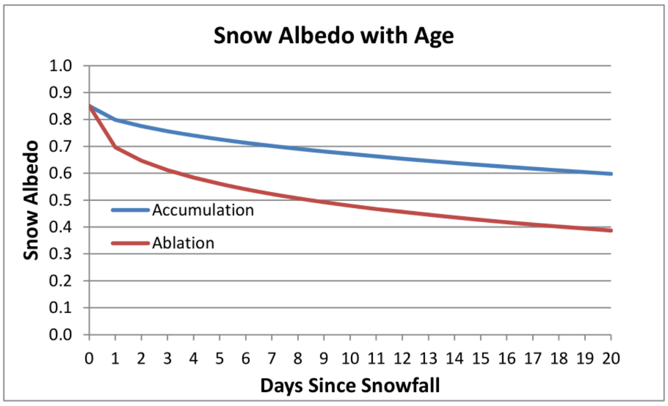

Figure 3. Relationship between snow albedo and snow age during accumulation and ablation periods (Ramboll., 2020).

Q3: The OH concentration differences mentioned at line 460 seem large (OH concentration difference ranged from -(1.73 × 10^6) cm^{-3} to 52.22 ×10^6 cm^{-3}) and I suggest double checking.

Response:

We sincerely appreciate the reviewer's in-depth discussion regarding the simulated concentration level of oxidative radical. We fully agree that the typical atmospheric OH concentration is on the order of 10^5–10^7 cm^{-3} (Holland et al., 2003; Yu et al., 2024). To address this discrepancy, we initially suspected potential sources of error such as incorrect parameterizations of OH-related chemical reactions or issues related to mass conservation in the chemical mechanism. However, these possibilities were ruled out after thorough rechecking. Subsequent investigation identified the root cause as a coding issue in the CAMx model: the simulated OH concentrations were mislabeled in the output NetCDF files. Specifically, while the model output indicated a unit of μg/m³, our analysis confirmed that the actual unit used in the model's source code was ppm (volume mixing ratio). This mislabeling led to an erroneous conversion when translating OH concentrations from μg/m^3 to cm^{-3}.

More details as follows:

1) **Verification of Unit Conversion in CAMx Postprocessing**

This study employed CAMx V7.10 for chemical transport modeling, with OH concentration output units in μg/m^3 (Figure 4). After unit conversion (μg/m^3 to cm^{-3}), the OH concentration range was 10^3–10^4 cm^{-3}. We confirmed that there were no errors in the conversion from μg/m^3 to cm^{-3}, as detailed below:

To convert the atmospheric OH concentration from **μg/m³** to **molecules/cm³**, follow these steps:

Step 1: Convert μg to grams

1μg=10^{-6}g g

So, **OH concentration in g/m³** = Concentration in μg/m^3 × 10^{-6}

Step 2: Convert grams to moles (using molar mass of OH)

- Molar mass of OH (Oxygen + Hydrogen) = 16 (O) + 1 (H) =**17 g/mol**
- Moles of OH per m^3 = (Concentration in g/m^3) / (17 g/mol)

Step 3: Convert moles to molecules

- Avogadro's number = 6.022×10^{23} molecules/mol
- Molecules of OH per m^3 = (Moles of OH/m^3) × (6.022×10^{23} molecules/mol)

Step 4: Convert m^3 to cm^3

$1\ m^3 = 10^6\ cm^3$

- Molecules of OH per cm^3 = (Molecules of OH/m^3) / 10^6

Final formula:

$$OH(molecules\ cm^{-3}) = \left(\frac{OH(\mu g\ m^{-3})}{17}\right) \times 6.022 \times 10^{23} \times 10^{-6}$$

$$OH(molecules\ cm^{-3}) = OH(\mu g\ m^{-3}) \times 3.54 \times 10^{10}$$

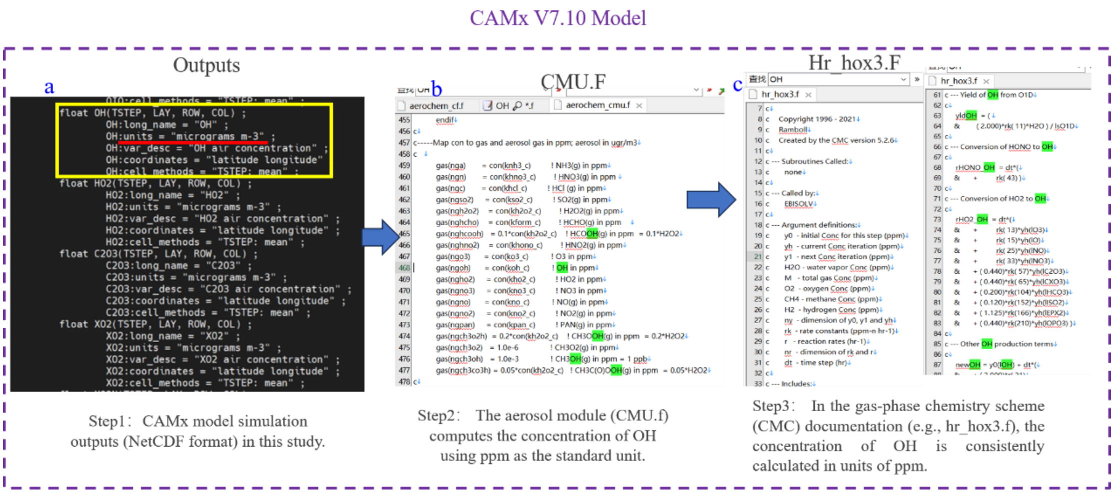

Figure 4. Unit analysis of OH concentration in CAMx codes.

2) Rechecking OH-related chemistry mechanism and the output module in CAMx

The CAMx is an open-source model, and the newly added N_2O_5 heterogeneous chemistry mechanism in this study was developed by our team and checked by Prof. Greg Yarwood (acknowledged in the paper) whom the major model developer of CAMx. Upon inspecting the source code, we found:

- OH is internally calculated in ppm in both aerosol (CMU.F) and gas-phase chemistry schemes (Figure 1b)
- The output module (IO_NCF) does not include a unit conversion step from ppm to $\mu g/m^3$ for certain gaseous species, such OH, RO_2, and HO_2. Thus, the "$\mu g/m^3$" unit labeled in the CAMx output files is incorrect, and the actual unit should be ppm.

3) Further cross-validation with outputted results from the CMAQ model

To further validate this issue, we analyzed OH concentrations simulated by the CMAQ model (which previously conducted by our research) over the same region. CMAQ outputs OH in ppbv (Figure 5). After conversion, **the concentration of OH can reach a range of** 10^5 to 10^7 cm^{-3}, supporting the conclusion that our CAMx-simulated OH concentrations fall within the expected range, and the discrepancy stemmed solely from unit mislabeling. These results are also in good agreement with previously observed OH concentration ranges (Holland et al., 2003; Yu et al., 2024).

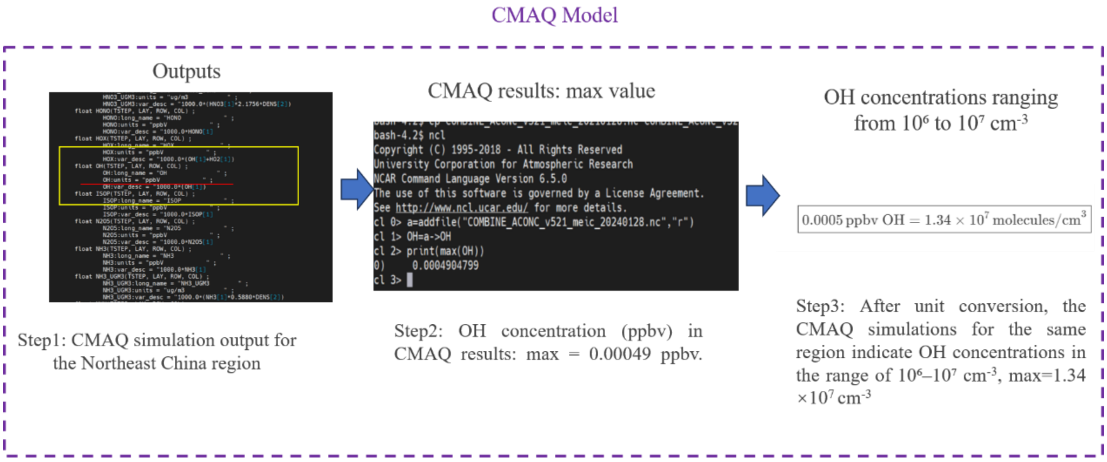

Figure 5. The concentration of OH simulated by CMAQ in the same region.

All in all, a coding error was identified in the official release of the CAMx model regarding the unit labeling of certain gaseous species in the output files: the OH, RO_2, and HO_2 concentration in the NetCDF output were incorrectly labeled as μg/m^3 (mass concentration), whereas the actual unit was ppm (volume mixing ratio). Accordingly, we have replotted Figures 7 and S4. Following thesuggestion of Reviewer 2, Figure 7 has been moved to theAppendix.

Reference：

Holland F, Hofzumahaus A, Schäfer J, et al. Measurements of OH and HO2 radical concentrations and photolysis frequencies during BERLIOZ[J]. Journal of Geophysical Research: Atmospheres, 2003, 108(D4): PHO 2-1-PHO 2-23.

Yu W, Zheng X, Tan M, et al. Field quantification of hydroxyl radicals by flow-injection chemiluminescence analysis with a portable device[J]. Environmental Science & Technology, 2024, 58(6): 2808-2816.

Q4: A suitable reference for the CAMx surface model is Karamchandani et al. (2015).

A suitable reference for the CAMx model is Emery et al., (2024). The CAMx v7.1 User'

s Guide could be cited (Ramboll, 2021).

Response: Thank you for these helpful literature suggestions. In the revised manuscript, we have added the following references at the relevant positions in the text:

- Karamchandani et al. (2015) has been cited as a reference for the CAMx surface model (Line 183).
- Emery et al. (2024) has been cited as a reference for the CAMx model (Line 114).
- Ramboll (2021), the CAMx v7.1 User's Guide, has also been cited (Line 114).

These references have been incorporated into the manuscript at the locations indicated above. We appreciate your careful guidance.

Q5: Is Bo Sea the same as the Bohai Sea? I think Bohai Sea is more commonly seen in English.
Response: Thank you for pointing this out. Yes, “Bo Sea” and “Bohai Sea” refer to the same body of water, i.e., the Bohai Sea in northern China. We agree with your comment that “Bohai Sea” is the more commonly used and standard term in English literature. Therefore, we have revised the manuscript to replace all instances of “Bo Sea” with “Bohai Sea”. We appreciate your careful observation.

Q6: In several places numeric values are given with more precision than needed, for example line 47 “contributing approximately 28.36% to nighttime accumulation” could be “approximately 28%”. Consider whether less precision would make numbers more readable.
Response: Thank you for the suggestion. We agree that excessive numerical precision can reduce readability and is often unnecessary. Accordingly, we will review all numeric values presented in the manuscript and adjust figures such as “28.36%” to a more appropriate level of precision (e.g., “approximately 28%”), thereby improving clarity while maintaining scientific accuracy. We appreciate your careful observation.

Q7: I found Figure S1 difficult to read, can the resolution be improved?
Response: Thank you for your comment. We agree that the resolution of Figure S1 is indeed too low, which affects its readability. In the revised manuscript, we will provide a higher-resolution and clearer version of Figure S1 to ensure all details are easily discernible. We appreciate you bringing this to our attention.

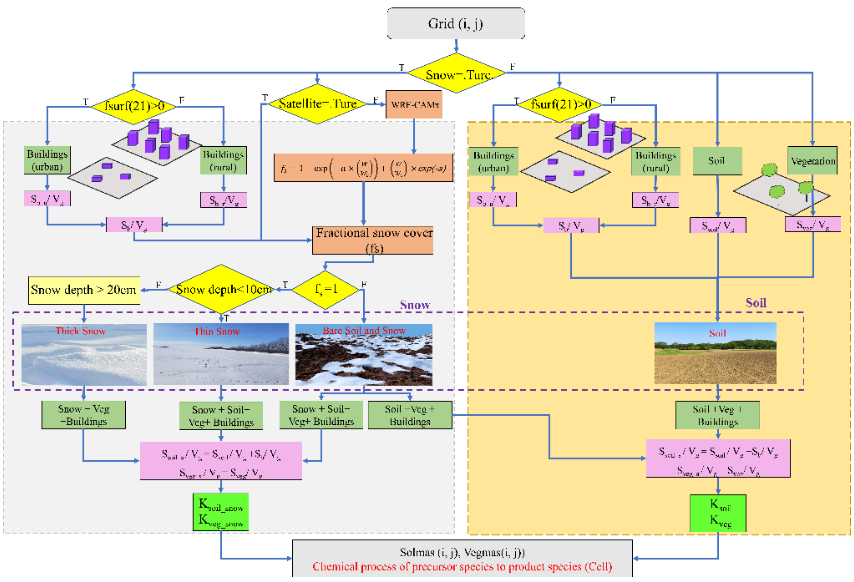

Figure S1. Characterization of S/V_g under different LU-LC.

References

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Chen, Q., et al., 2019. HONO, particulate nitrite, and snow nitrite at a midlatitude urban site during wintertime. ACS Earth and Space Chemistry. 3, 811-822.

Emery, C., et al., 2024. Comprehensive air quality model with extensions: formulation and evaluation for ozone and particulate matter over the US. Atmosphere, 15(10), p.1158.

Holland F, Hofzumahaus A, Schäfer J, et al. Measurements of OH and HO2 radical concentrations and photolysis frequencies during BERLIOZ[J]. Journal of Geophysical Research: Atmospheres, 2003, 108(D4): PHO 2-1-PHO 2-23.

Karamchandani, P., et al., 2015. Implementation and refinement of a surface model for heterogeneous HONO formation in a 3-D chemical transport model. Atmospheric Environment, 112, pp.356-368.

Fu, X., et al., 2019. The significant contribution of HONO to secondary pollutants during a severe winter pollution event in southern China. Atmospheric Chemistry and Physics. 19, 1-14.

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Ramboll, 2020. User's guide, comprehensive air quality model with extensions, version 7.10. Available at: https://www.camx.com/Files/CAMxUsersGuide_v7.10.pdf.