Implications of using MERRA-2 cloud water in aerosol wet scavenging for Pb-210 surface concentrations and vertical profiles in GEOS-Chem

Bo ZHANG[1] (bo.zhang@nianet.org), Hongyu LIU[1] (hongyu.liu-1@nasa.gov), James H. CRAWFORD[2], T. Duncan FAIRLIE[2], Gao CHEN[2],
Jack E. Dibb[3], Viral SHAH[4], Gan Luo[5], Melissa P. SULPRIZIO[4], and Robert M. YANTOSCA[4]
[1] National Institute of Aerospace, Hampton, VA, USA, [2] NASA Langley Research Center, Hampton, VA, USA [3] University of New Hampshire, Durham, NH, USA,
[4] Harvard University, Cambridge, MA, USA [5] State University of New York, Albany, NY, USA

Introduction

Precipitation scavenging is the dominant loss process for a whole suite of aerosols but model parameterizations of this process are highly uncertain, substantially contributing to large uncertainties in the simulated loadings and radiative forcing of aerosols. Lead-210 (^{210}Pb, radioactive half-life of 22.3 years) is produced by radioactive decay of soil-emitted gaseous ^{222}Rn. It attaches to ambient submicron aerosols and is subject to precipitation scavenging processes. Liu et al. [2001] estimated the global mean lifetime of tropospheric ^{210}Pb aerosols to be ~9 days using the GEOS-Chem model. Luo et al. [2019, 2020] provided an option to use MERRA-2 cloud water in the model aerosol scavenging scheme. In this study, we examine the impact of this revision on ^{210}Pb simulations and compare results with climatological observations of ^{210}Pb surface and UT/LS concentrations, as well as aircraft profile measurements from eleven NASA field missions.

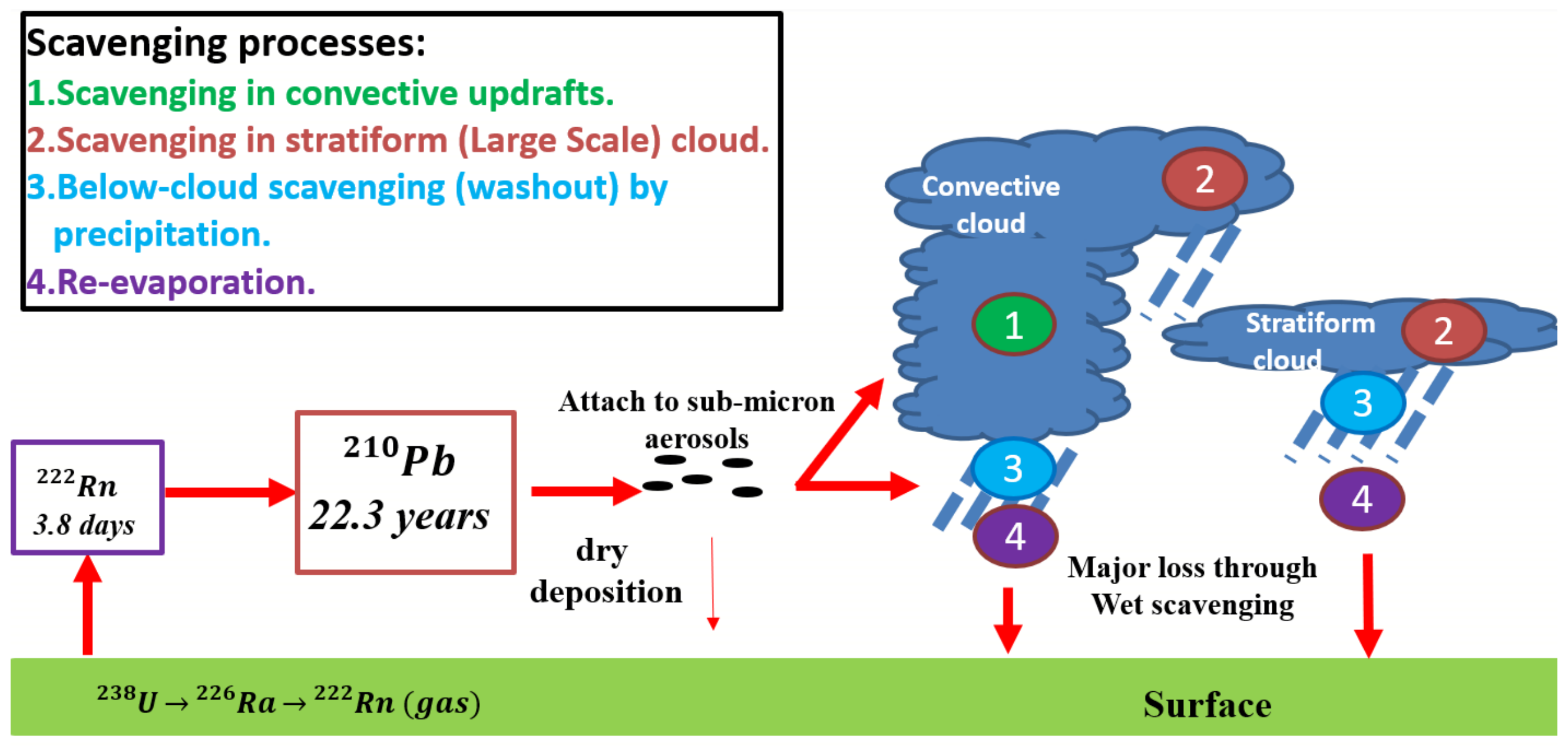


Figure 1. Schematic showing the processes determining ^{210}Pb in the troposphere. Aerosol scavenging processes in GEOS-Chem are numbered and stated in the legend. Wet deposition is the principal sink of ^{210}Pb in the troposphere.

Default aerosol rainout:

$$F = f(1 - e^{-ak\Delta t})$$

- F is aerosol fraction removed by scavenging; f is cloud fraction.
- k is the rate of conversion from cloud water to precipitation. a is a temperature (T) dependent coefficient, given in the form of a 3-element vector representing the partitioning of aerosol particles into cloud droplets or ice crystals at T< 237 K, 237 K ≤ T < 258 K, and T ≥ 258 K.
- Partitioning of 100% ^{210}Pb into ice (T< 237 K).

Revised by Luo et al. [2019 & 2020]

$$F = \frac{P_r}{k \cdot \mathrm{ICCW}}\left(1 - e^{-k\cdot\Delta t}\right) = \frac{f_c \cdot P_r}{k\left(\mathrm{LCW} + \mathrm{ICW} + P_r \cdot \Delta t\right)}\left(1 - e^{-k\cdot\Delta t}\right)$$

- Use the sum of MERRA-2 non-precipitating cloud water content (LCW+ICW) and precipitating cloud water (PΔt) for aerosol scavenging.
- Use cloud fraction to convert in-cloud LCW and ICW to grid average values.
- Partitioning of 40% ^{210}Pb into ice (T< 237 K).

Model and Data

GEOS-Chem

- v11-01 driven by MERRA-2. MERRA-2 variables involved in cloud scavenging: precipitation formation rate, precipitation flux, precipitation evaporation, liquid/ice cloud water content, cloud fraction, cloud mass flux, and convective entrainment.
- 2°× 2.5° horizontal resolution, 72 vertical levels.
- ^{222}Rn-^{210}Pb-^{7}Be simulation: ^{222}Rn emissions from Jacob et al. [1990] or B. Zhang et al. [2021].

^{210}Pb Observations

- Surface ^{210}Pb observational data compiled by Preiss et al. [1996] & F. Zhang et al. [2021].
- **RANDAB:** a radionuclide database compiled from high-altitude aircraft and bolloon measurements conducted during 1950s-1980s. It has been used to evaluate ^{210}Pb in the upper troposphere and lower stratosphere (UT/LS) in global models.
- NASA aircraft campaigns:

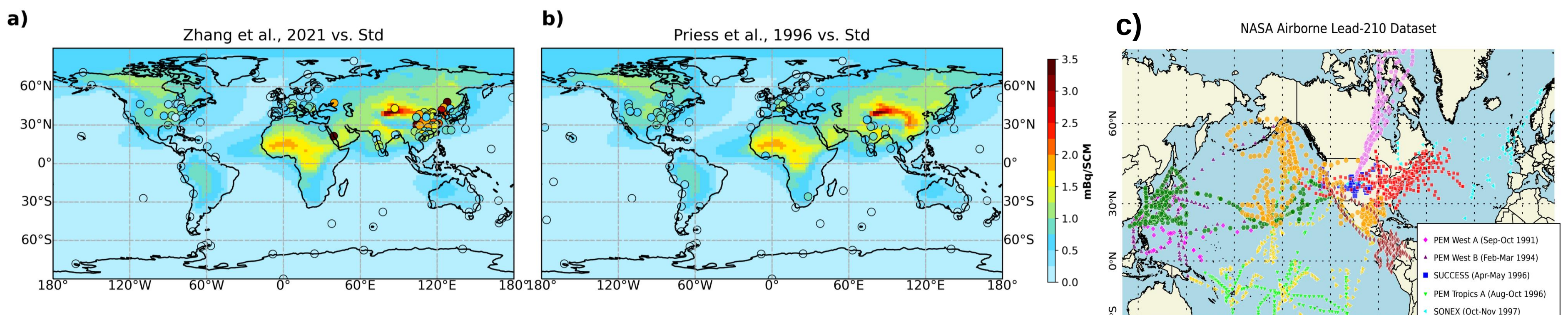


Figure 2. Annual mean surface observations (color-coded circles) of ^{210}Pb collected by a) F. Zhang et al. [2021] and b) Preiss et al. [1996] in comparison with GEOS-Chem results (color image). c) Eleven NASA aircraft campaigns with ^{210}Pb vertical profile measurements.

Satellite cloud water measurements

2B-CWC-RO (cloud liquid water content or CLWC based on CloudSat Radar-Only) and 2C-ICE (cloud ice water content or CIWC based on CloudSat and CALIPSO) obtained from the CERES Level-2 CCCM product [Kato et al., 2010]. We sampled co-located CWC in MERRA-2 (3-hour average, 0.5×0.625°) along the satellite orbit track for comparison.

Results and Discussion

Experiments

Model experiments with different wet scavenging scheme configurations are performed to test:

- Two ^{222}Rn emission options (Jacob et al., 1997 vs. B. Zhang et al., 2021): JA97 vs. Std.
- Adding scavenging in mixed-phase clouds (no scavenging by default): MixPhase05.
- New scavenging option using CWC in MERRA-2 as suggested by Luo et al. [2019, 2020]: Luo20
- Reduced ice cloud scavenging based on Luo et al. [2019, 2020]: Luo20_NoIceScav, Luo20_20PercIceScav.
- Using monthly averaged lower-tropospheric CWC from MERRA-2 instead of spatiotemporally varying values: CWC-L-O-Mon.

Comparisons with ^{210}Pb at surface (Preiss et al., 1996 and F. Zhang et al., 2021) and UT/LS (RANDAB)

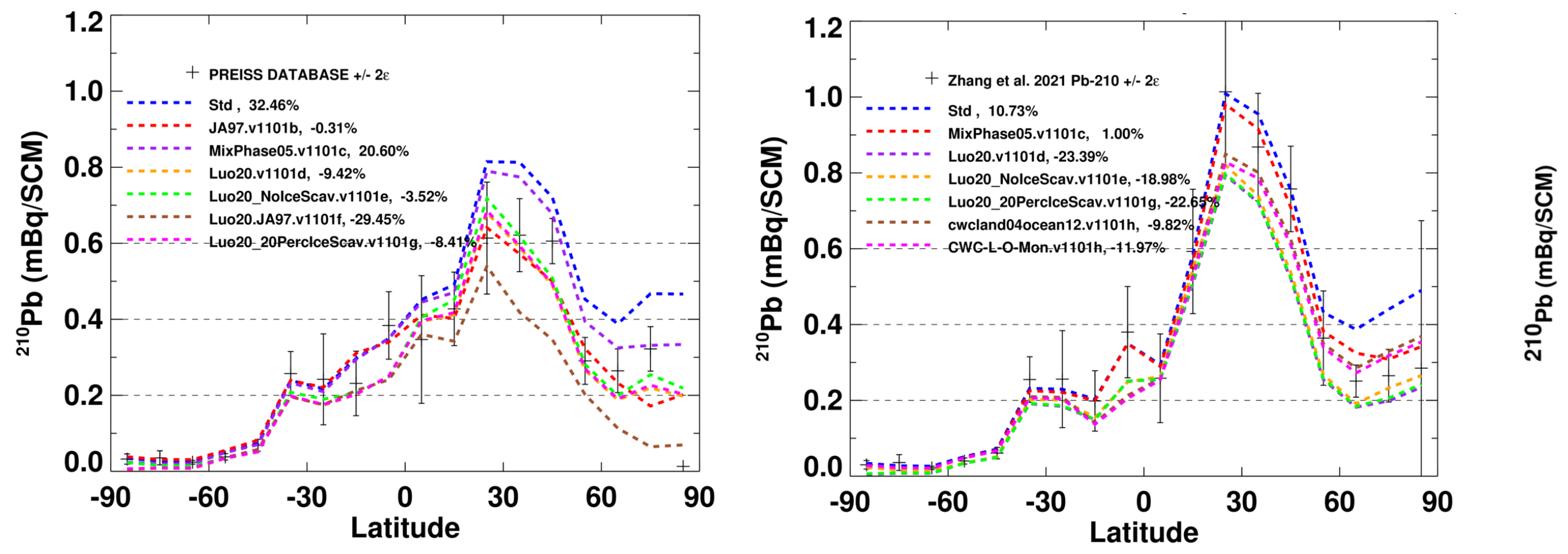


Figure 3. Comparisons of observed and simulated latitudinal distributions of annually averaged ^{210}Pb concentrations at surface. The observed distribution is calculated by averaging observations from Preiss et al. (1996) (left) and F. Zhang et al. (2021) (right). Error bars represent ±2 times the standard error of the averages.

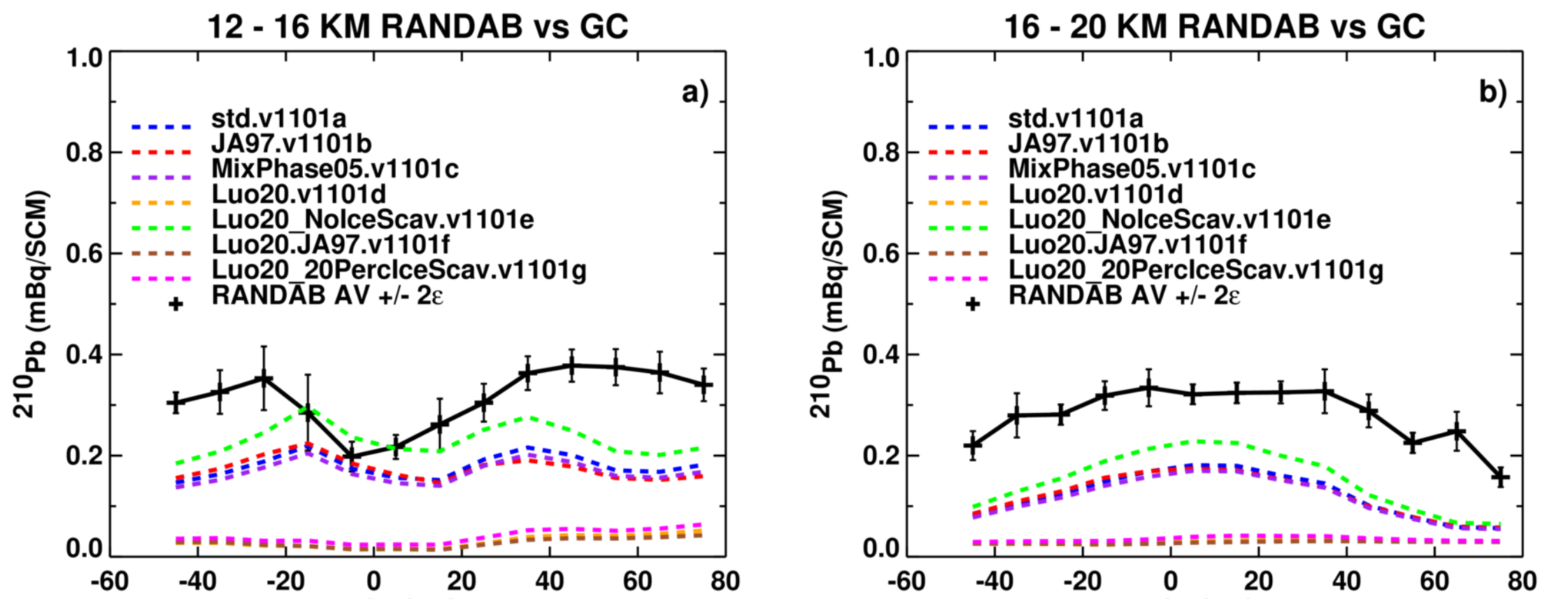
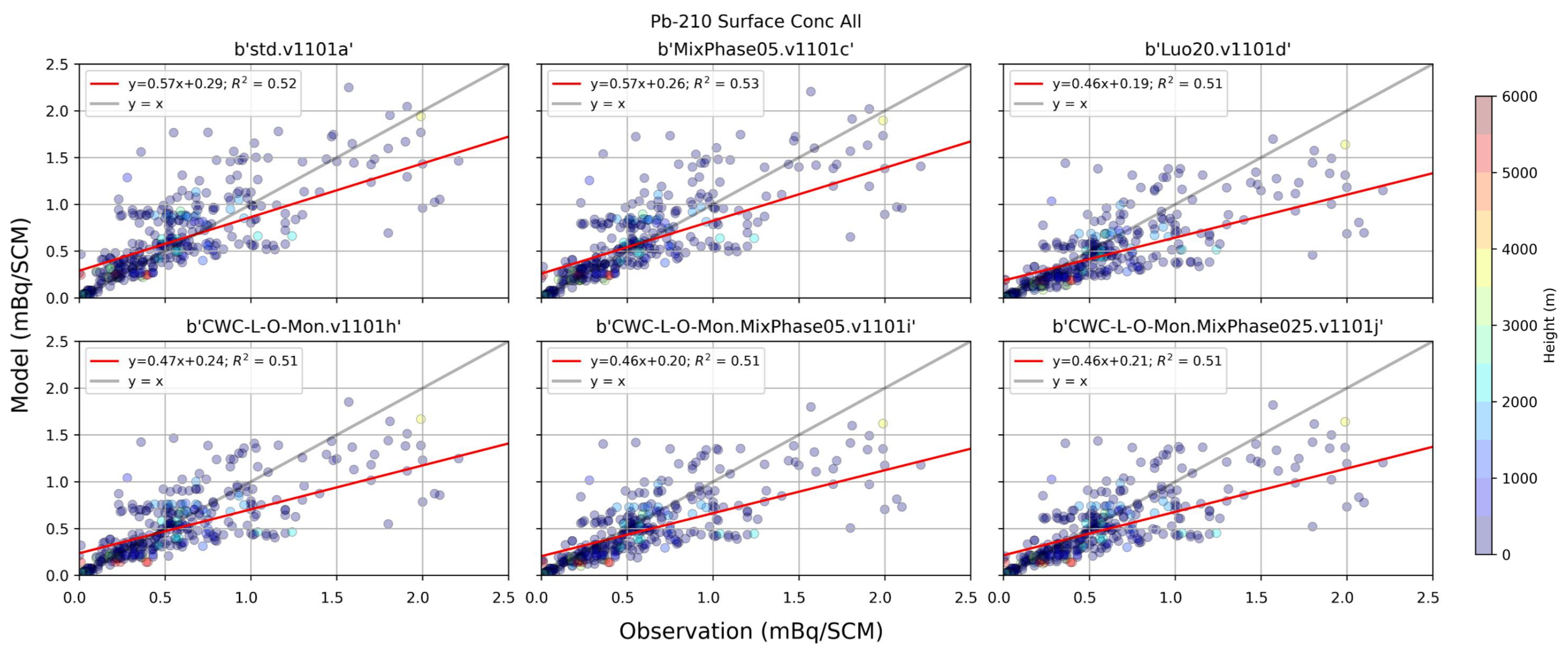


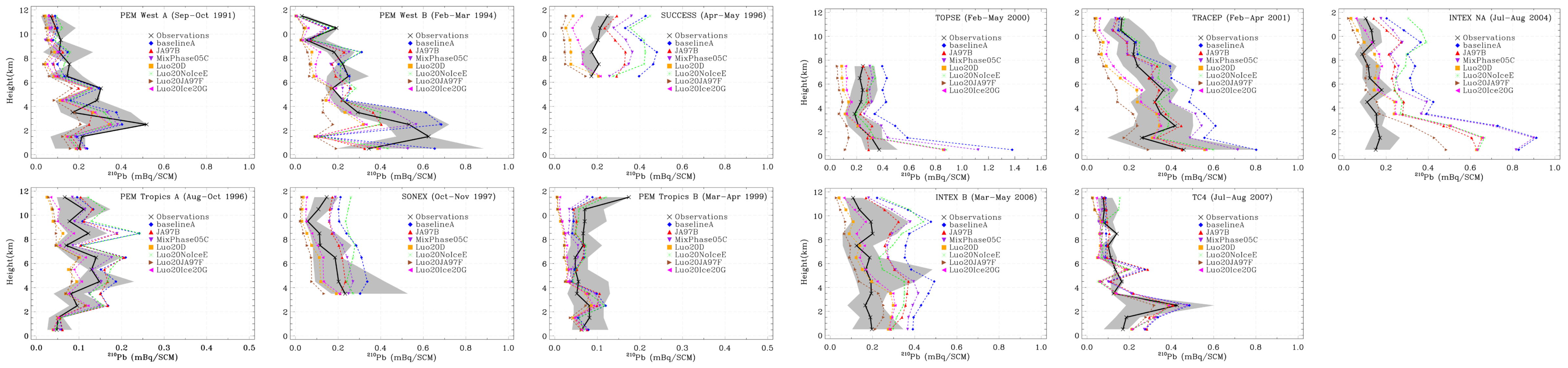
Figure 4. Comparisons of observed and simulated latitudinal distributions of annually averaged ^{210}Pb concentrations in UT/LS (left for 12–16 km altitudes and right for 16–20 km altitudes). Observations from the US Environmental Measurement Laboratory RANDAB database are averaged into 10° latitude bins.

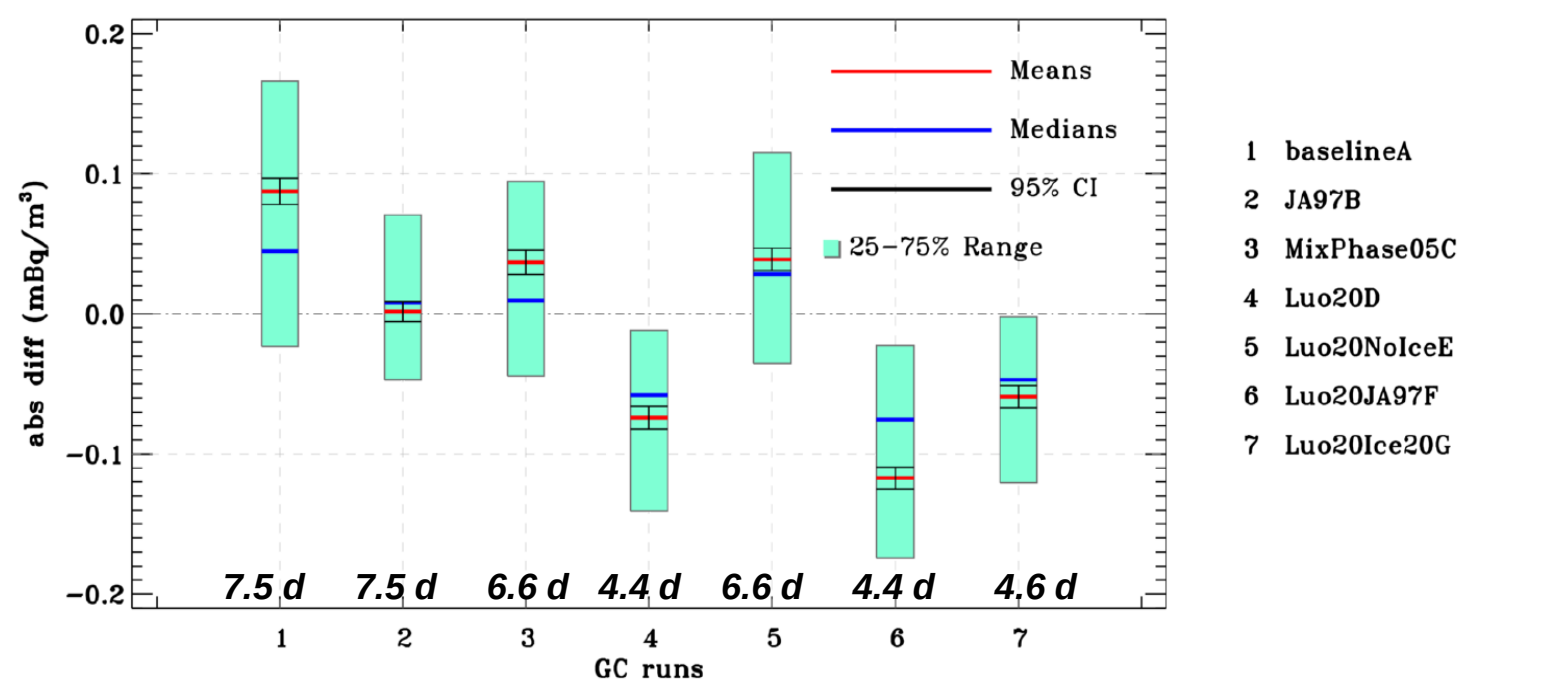


← Figure 5. Scatterplots comparing simulated ^{210}Pb surface concentrations with observations from F. Zhang et al. (2021) for each model experiment.

- The new wet scavenging option [Luo et al., 2020] with the original ^{222}Rn emission [Jacob et al., 1997] leads to large underestimates of ^{210}Pb surface concentrations.
- With the B. Zhang et al. [2021] ^{222}Rn emission option, ^{210}Pb surface concentrations are still underestimated at surface and in the UT/LS.

Comparisons with NASA aircraft ^{210}Pb profiles





↑ Figure 6. Comparisons between simulated and observed ^{210}Pb profiles for NASA campaigns. The black dots show the individual measurements. The solid black lines indicate the averaged concentrations in 1-km altitude bins. The color lines show the corresponding binned concentrations for the model experiments.

← Figure 7. The averaged differences between simulated ^{210}Pb and NASA aircraft measurements. The dashed-dotted horizontal line indicates zero difference. The statistics and sensitivity simulations are given in the legends. Global mean lifetime of tropospheric ^{210}Pb (days) for each experiment is also denoted.

Ice Cloud Water Content in MERRA-2 compared with CLIPSO-CloudSat ice cloud water

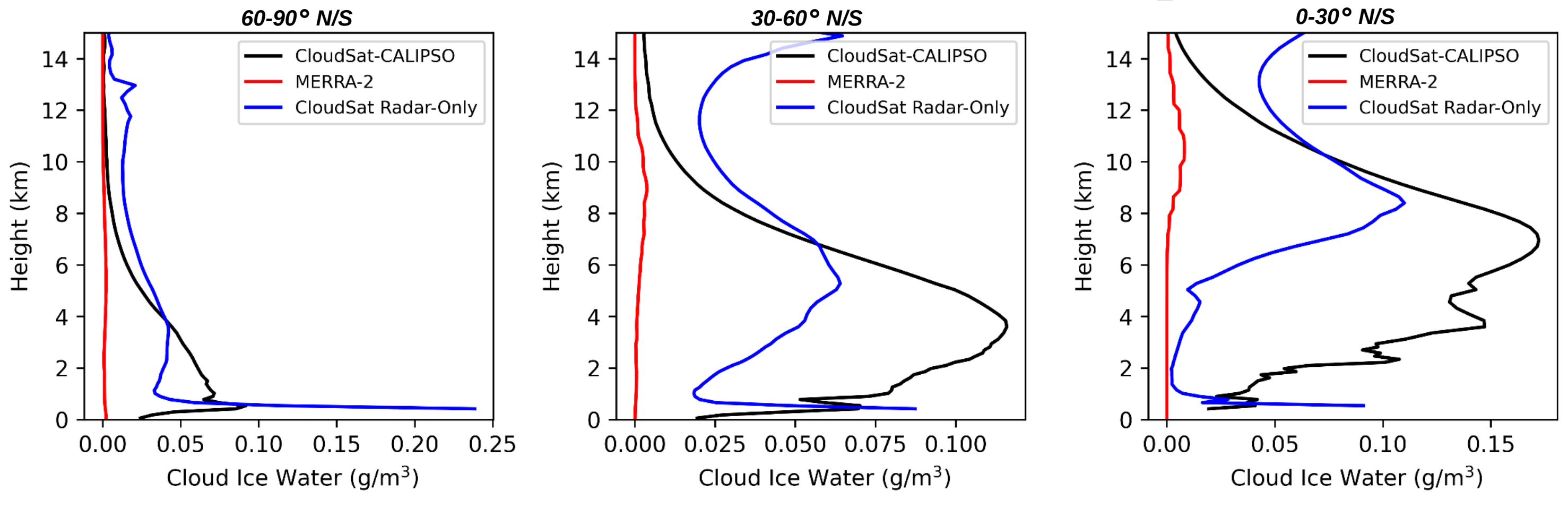
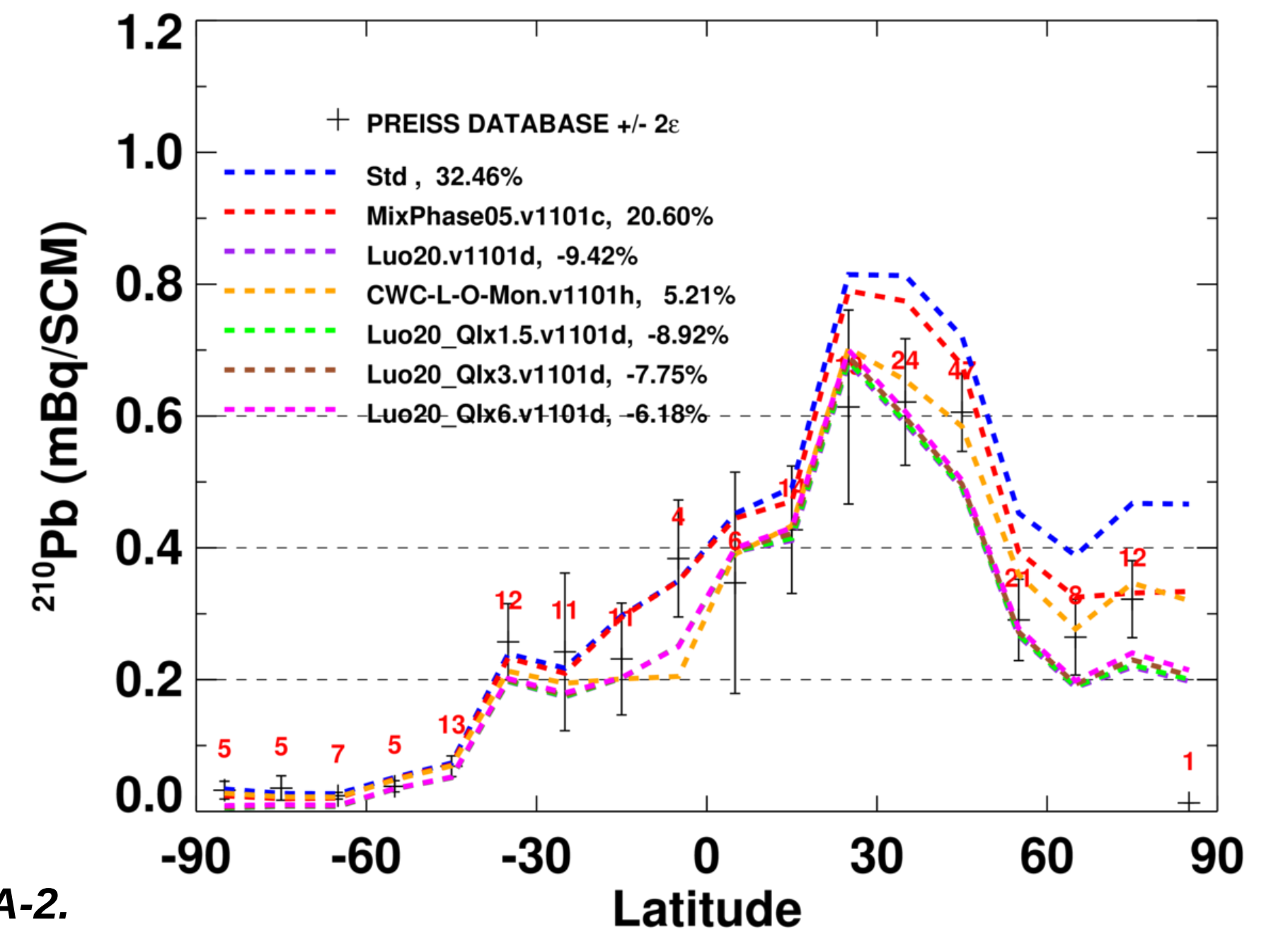


Figure 8. Annual zonal mean vertical profiles of co-located cloud ice water (g/m³) in 2C-ICE (black, Level-2), CloudSat Radar-Only (2B-CWC-RO; blue, Level-2), and MERRA-2 (red, 3-hour average) for the three latitude regions in 2008.



→ Figure 9. Same as Fig. 3 but for experiments with modified (enhanced) ice cloud water in MERRA-2.

Conclusions

- NASA aircraft ^{210}Pb profile measurements provide strong constraints on model aerosol scavenging and thus global mean lifetime of tropospheric ^{210}Pb aerosols (~ 7 days).
- Using MERRA-2 cloud water content in the large-scale scavenging scheme [Luo et al., 2020] in GEOS-Chem leads to large underestimates of ^{210}Pb conc., especially in the mid-/upper- troposphere and at NH high-latitude surface. Reducing scavenging in ice clouds can significantly improve the model bias.
- MERRA-2 in-cloud ice water content is biased low compared to CLIPSO-CloudSat observations, suggesting issues with cloud water partitioning between liquid and ice in MERRA-2. Further improvement of the Luo et al. [2020] scavenging scheme that uses MERRA-2 cloud water requires evaluation of MERRA-2 cloud water content with available observations including satellite measurements.

References:

Kato, S., S. Sun-Mack, W.F. Miller, F.G. Rose, Y. Chen, P. Minnis, & B.A. Wielicki (2010): Relationships among cloud occurrence frequency, overlap, and effective thickness derived from CALIPSO and CloudSat merged cloud vertical profiles, J. Geophys. Res., 115:D00H28. doi:10.1029/2009JD012277.

Liu, H., Jacob, D. J., Bey, I., & Yantosca, R. M. (2001). Constraints from ^{210}Pb and ^{7}Be on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields. *Journal of Geophysical Research: Atmospheres*, *106*(D11), 12109-12128.

Luo, G., Yu, F., and Schwab, J. (2019): Revised treatment of wet scavenging processes dramatically improves GEOS-Chem 12.0.0 simulations of surface nitric acid, nitrate, and ammonium over the United States, Geosci. Model Dev., 12, 3439–3447, https://doi.org/10.5194/gmd-12-3439-2019.

Luo, G., Yu, F. and Moch, J.M. (2020): Further improvement of wet process treatments in GEOS-Chem v12. 6.0: impact on global distributions of aerosols and aerosol precursors. Geoscientific Model Development, 13(6), pp.2879-2903.

Zhang, B., Liu, H., Crawford, J. H., Chen, G., Fairlie, T. D., Chambers, S., Kang, C.-H., Williams, A. G., Zhang, K., Considine, D. B., Sulprizio, M. P., Yantosca, R. M. (2021). Simulation of radon-222 with the GEOS-Chem global model: emissions, seasonality, and convective transport. Atmospheric Chemistry and Physics, 21(3), 1861-1887.

Zhang, F., Wang, J., Baskaran, M., Zhong, Q., Wang, Y., Paatero, J., & Du, J. (2021). A comprehensive global dataset of atmospheric 7Be and 210Pb measurements: air concentration and depositional flux. Earth System Science Data, 7, 1-75.

Acknowledgement: This work is supported by NASA's ACCDAM program (NNX14AR07G and 80NSSC21K1455).