Constraints From Airborne ^{210}Pb Observations on Aerosol Scavenging and Lifetime in a Global Chemical Transport Model

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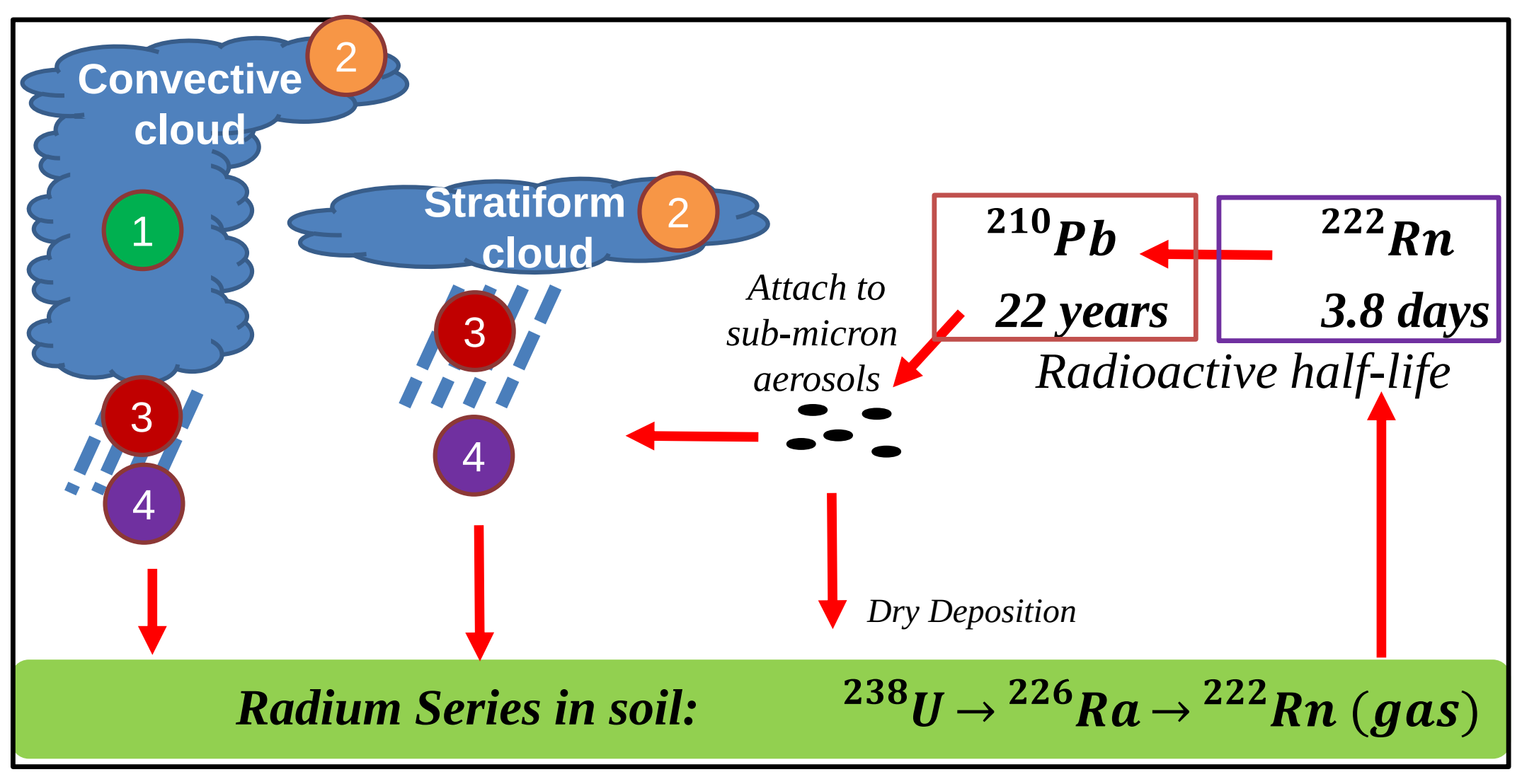
Introduction

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. More detailed treatments of precipitation scavenging processes (e.g., scavenging in ice and mixed-phase clouds) have recently been developed and applied to the model [Wang et al., 2011, 2014], and may alter the ^{210}Pb distribution and lifetime. In addition, NASA aircraft campaigns over the past two decades have provided substantial records of ^{210}Pb profiles around the world. In this study, we use these datasets to constrain aerosol scavenging parameterization in GEOS-Chem and to estimate observation-based ^{210}Pb aerosol lifetime.



1. Scavenging in convective updrafts;
2. In-cloud scavenging (rainout) by stratiform & convective anvil precipitation;
3. Below-cloud scavenging (washout) by precipitation;
4. Re-evaporation.

Figure 1. Life cycle of ^{210}Pb in the troposphere.

Parameterizations of precipitation scavenging

Assumed linear removal:
$$\frac{\Delta C_i}{\Delta t} = F \cdot ak_i \cdot C_i$$

- C_i is the mixing ratio of tracer i (e.g., ^{210}Pb);
- F is the areal fraction that actually experiences precipitation;
- k_i is the assembled scavenging coefficient accounting for various scavenging processes. a is a temperature (T) dependent coefficient, given in the form of a 3-element vector representing efficiencies for T< 237 K, 237 K <T < 258 K, and T > 258 K.

Table 1. Parameterizations of the scavenging coefficient k_i

	Convective Precipitation	**Stratiform Precipitation**
In-cloud scavenging (ICS)	k_i is determined by updraft velocity and a fixed conversion rate from cloud condensate to precipitation. ①	k_i accounts for conversion from cloud condensate to precipitation and cloud drop accretion process. ②
Below-cloud Scavenging (BCS)	$k_i = c_1 \times (\frac{P}{F})^{c_2}$ is the overall BCS coefficient determined for an assumed typical raindrop and aerosol sizes for impaction, interception and diffusion processes. $P =$ precipitation, $c1$ and $c2$ vary for aerosol size and temperature [Wang et al., 2011]. ③	
Re-evaporation	50% of aerosol is released back to ambient air for the amount of precipitation evaporated. ④	

Other related parameters:

- Cloud water content (CWC, having unit of $\frac{cm^3\ water}{cm^3\ air \cdot s}$). It is considered as a constant parameter, which defines water density of cloud. It consists of liquid water content and ice water content, and the allocation is temperature dependent. For a given rate of precipitation formation, increase in CWC reduces the fraction experiencing in-cloud scavenging (i.e., F).

Model and Data

GEOS-Chem

- v11-01 driven by MERRA. MERRA variables involved in cloud scavenging are new precipitation formation, precipitation flux, precipitation evaporation, cloud mass flux, entrainment in convective updraft.
- 2°× 2.5° horizontal resolution and 47 vertical levels.
- Rn-Pb-Be simulation option with Radon emission defined by Jacob et al. [1990].

^{210}Pb Observations

- Latitudinal surface ^{210}Pb distribution compiled by Preiss et al. [1996]
- **RANDAB** is a radionuclide database compiled from high-altitude aircraft and bolloon measurements conducted during 1950s-1980s. It has specifically been used to evaluate simulated ^{210}Pb in the upper troposphere and lower stratosphere (UT/LS).
- NASA aircraft campaigns:
  PEM-West A, PEM-West B, TRACE-P, PEM-Tropics A, PEM-Tropics B, SUCCESS
  SONEX, TOPSE, INTEX-NA, INTEX-B, TC4

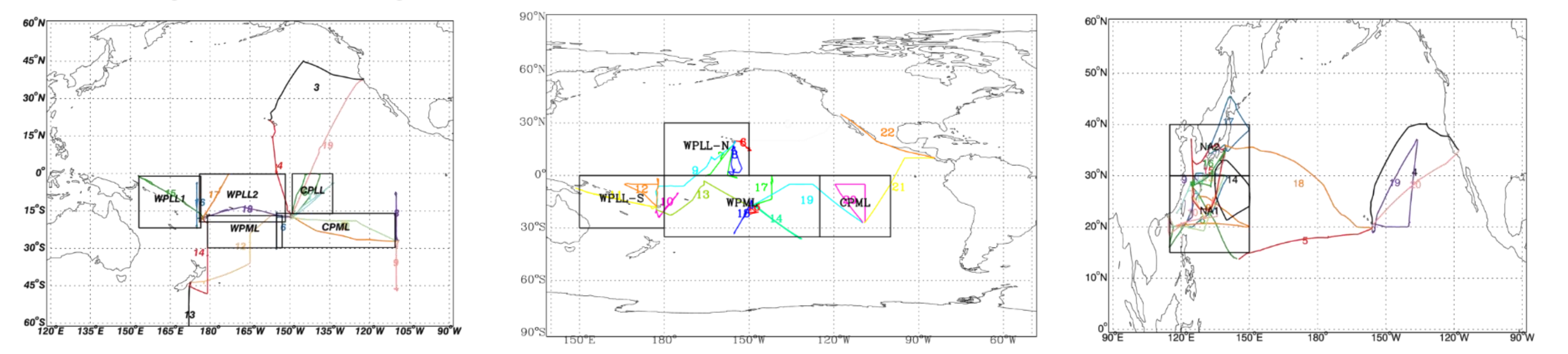


Figure 2. Flight tracks for three NASA aircraft campaigns investigated in this study. Black rectangles indicate groups of data for examining regional characteristics of precipitation scavenging of aerosols.

Results and Discussion

Experiments

Table 1. Settings of experiments & features of simulated ^{210}Pb

Experiment	**Details**	**Trop ^{210}Pb lifetime (days)**	**Annual zonal mean (Fig. 3)**	**Comparison with Preiss surface ^{210}Pb and RANDAB (Fig. 4)**	**Comparison with ^{210}Pb profiles (Fig. 5)**
Standard	CWC = 1.0 $\frac{cm^3\ water}{cm^3\ air \cdot s}$ $\boldsymbol{a}$ (convective) = 1.0-0.5-1.0 $\boldsymbol{a}$ (stratiform) = 1.0-0.0-1.0	8.32	Highest conc near the middle latitudinal surface; low conc in the tropical MT/UT due to efficient scavenging.	Overestimate at surface compared with Preiss data; underestimate in UT/LS.	Compared well with TRACE-P. Moderate overestimate for PEM-West A in UT and above PBL for PEM-West B.
CWC15	Same as Std, but CWC = 1.5 $\frac{cm^3\ water}{cm^3\ air \cdot s}$	9.49	Significant increase throughout the troposphere at mid/high latitudes.	Mostly overestimate	Overestimate in most levels in the troposphere.
cvrain000510	Same as Std, but $\boldsymbol{a}$ (convective) = 0.0-0.5-1.0	8.36	Barely noticeable increase across the tropical tropopause.	Barely noticeable changes.	Barely noticeable changes.
lsrain000010	Same as Std, but $\boldsymbol{a}$ (stratiform) = 0.0-0.0-1.0	9.37	Large increase across the tropical tropopause	Best agreement with UT/LS obs.	Too much ^{210}Pb in UT.
lsrain100510	Same as Std, but $\boldsymbol{a}$ (stratiform) = 1.0-0.5-1.0	7.41	Remarkable reduction in the middle/upper troposphere (MT/UT)	Best agreement with surface meridional distribution.	Similarly decent performance with some extent of underestimate.

Simulated annual zonal mean

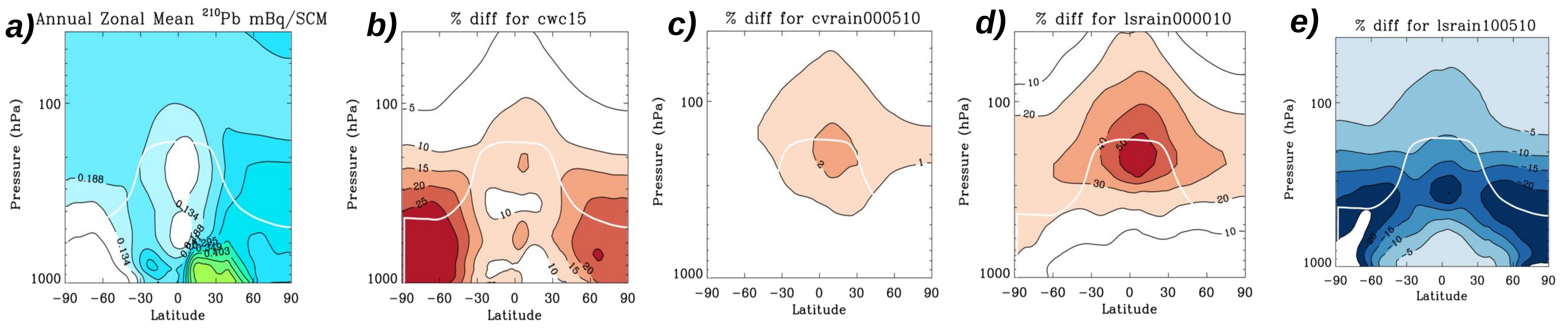


Figure 3. Simulated annual zonal mean ^{210}Pb in the standard setting of GEOS-Chem (a) and the percentile differences ($\frac{Exp - Std}{Std}$) as a result of changes in scavenging parameterizations: CWC15 (b), cvrain000510 (c), lsrain000010 (d), and lsrain100510 (e).

Comparison with ^{210}Pb obs. @ surface (Preiss et al.) and UT/LS (RANDAB)

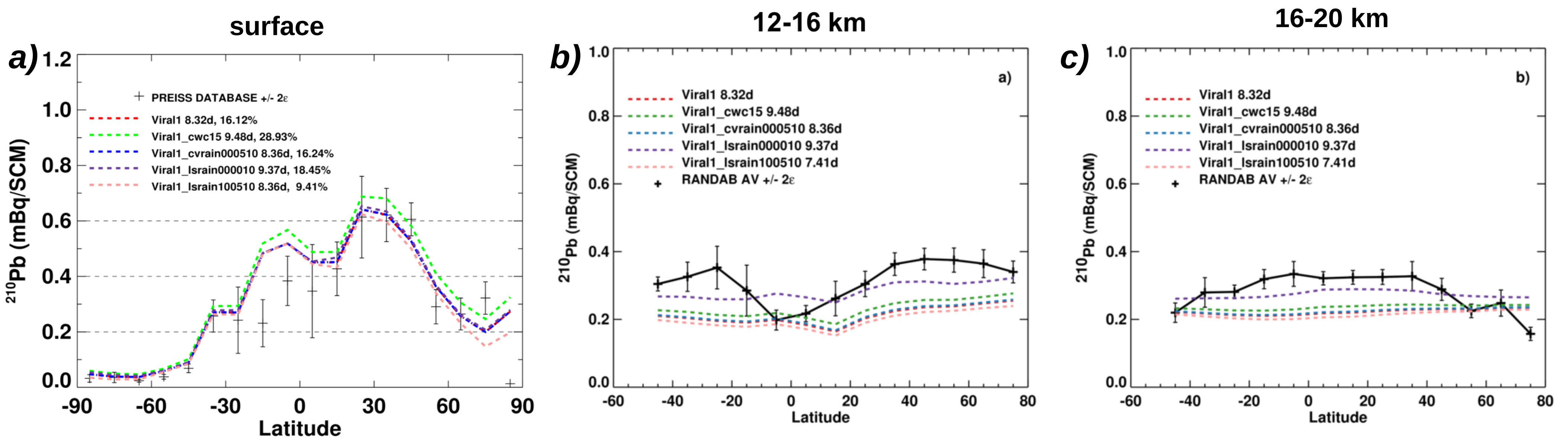


Figure 4. Comparisons of observed and simulated latitudinal distributions of annually averaged ^{210}Pb concentrations at surface (a) and UT/LS ((b) for 12–16 km level and (c) for 16–20 km level). The observed distribution is calculated by averaging observations from the Preiss et al. (1996) database and the US Environmental Measurement Laboratory RANDAB database into 10° latitude bins. Error bars represent ±2 times the standard error of the averages. Simulated distributions were obtained by sampling model output at observation locations and then treating model output in the same manner as the observations.

Comparison with ^{210}Pb profiles

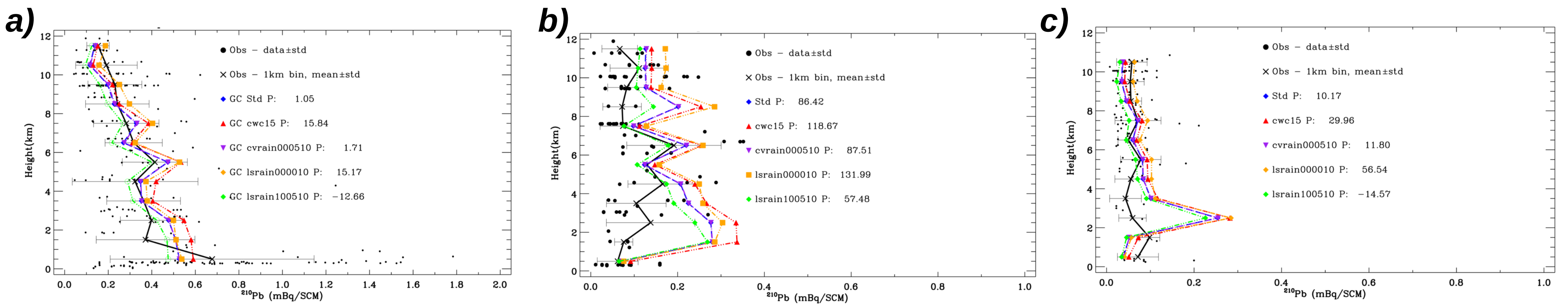


Figure 5. Comparisons of observed and simulated ^{210}Pb profiles during three NASA aircraft campaigns: (a) TRACE-P, (b) PEM-West A, and (c) PEM-West B. P values indicate the overall percentile differences between simulated results and observations.

Conclusions

- Lead-210 distribution and lifetime in the atmosphere are not sensitive to ice in-cloud scavenging in convective updraft. Ice in-cloud scavenging in stratiform clouds reduces tropospheric ^{210}Pb lifetime by ~ 1 day and results in better agreements with observed surface observations and aircraft measured profiles. However, the process results in significant underestimate of ^{210}Pb in UT/LS.
- Increase in cloud water content by 50% leads to an increase of ^{210}Pb lifetime by ~ 1 day, largely due to the increase in ^{210}Pb concentrations at mid/high latitudes.
- Mixed-phase in-cloud scavenging for stratiform clouds has a reducing impact on the ^{210}Pb lifetime by ~ 1 day. Results match better with the Preiss surface observations and aircraft profiles. This suggests that such process (i.e., impaction) needs to be incorporated in models.
- Comparisons with NASA aircraft ^{210}Pb profiles suggest the estimated tropospheric ^{210}Pb lifetime should be close to 7.4-8.3 days. Further analyses against the rest of aircraft campaigns will provide a better constraint on the estimate.

Future work

- Determine the sensitivity of simulated ^{210}Pb in different regions / latitudes to changes in cloud scavenging parameters;
- Adjust parameterizations based on current findings to better match NASA aircraft observations;
- Obtain a global mean ^{210}Pb lifetime constrained by all NASA aircraft campaigns.

References:

Jacob, D. J., & Prather, M. J. (1990). Radon-222 as a test of convective transport in a general circulation model. *Tellus B*, *42*(1), 118-134.

Preiss, N., Mélières, M. A., & Pourchet, M. (1996). A compilation of data on lead 210 concentration in surface air and fluxes at the air-surface and water-sediment interfaces. *Journal of Geophysical Research: Atmospheres*,*101*(D22), 28847-28862.

Liu, H., Jacob, D. J., Bey, I., & Yantosca, R. M. (2001). Constraints from 210Pb and 7Be 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.

Wang, Q., Jacob, D. J., Fisher, J. A., Mao, J., Leibensperger, E. M., Carouge, C. C., ... & Doherty, S. J. (2011). Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: implications for radiative forcing. *Atmospheric Chemistry and Physics*,*11*(23), 12453-12473.

Wang, Q., Jacob, D. J., Spackman, J. R., Perring, A. E., Schwarz, J. P., Moteki, N., ... & Barrett, S. R. (2014). Global budget and radiative forcing of black carbon aerosol: Constraints from pole-to-pole (HIPPO) observations across the Pacific. *Journal of Geophysical Research: Atmospheres*, *119*(1), 195-206.

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