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 (210Pb), radioactive half-life of 22.3 years is produced by radioactive decay of soil-emitted gases. It attaches to ambient submicron aerosols and is subject to precipitation scavenging processes. Liu et al. [2001] estimated the global mean lifetime of tropospheric 210Pb 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 210Pb distribution and lifetime. In addition, NASA aircraft campaigns over the past two decades have provided substantial records of 210Pb profiles around the world. In this study, we use these datasets to constrain aerosol scavenging parameterization in GEOS-Chem and to estimate observation-based 210Pb aerosol lifetime.

Experiments

Results and Discussion

Simulated annual zonal mean

Comparison with 210Pb obs. @ surface (Preiss et al.) and UTLS (RANDAB)

Figure 4. Comparisons of observed and simulated latitudinal distributions of annually averaged 210Pb concentrations at surface (a) and UTLS (b) for 12–15 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 U.S. Environmental Monitoring Laboratory RANDAB database into 10° latitude bins. Error bars represent ±2 times the standard error of the averages. Simulated distributions were obtained from sampling model output at observation locations and then treating model output in the same manner as the observations.

Comparison with 210Pb profiles

Figure 5. Comparisons of observed and simulated 210Pb profiles during three NASA aircraft campaigns: (a) TRACE-P, (b) PEM-West A, and (c) PEM-West B. 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 210Pb lifetime by ~1 day and results in better agreements with observed surface observations and aircraft measured profiles. However, the process results in significant underestimation of 210Pb in UTLS.
- Increase in cloud water content by 50% leads to an increase of 210Pb lifetime by ~1 day, largely due to the increase in 210Pb concentrations at mid/high latitudes.
- Mixed-phase ice-in-cloud scavenging for stratiform clouds has a reducing impact on the 210Pb lifetime by ~1 day. Results match better with the Press surface observations and aircraft profiles. This suggests that such process (i.e., impaction) needs to be incorporated in models.
- Comparisons with NASA aircraft 210Pb profiles suggest the estimated tropospheric 210Pb 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 210Pb 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 210Pb lifetime constrained by all NASA aircraft campaigns.

Acknowledgement: This study is funded by NASA Aeronautics program (NNIN218009A).

References:

[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] University of Washington, Seattle, WA, USA.
[5] Harvard University, Cambridge, MA, USA.

Parameterizations of precipitation scavenging

Assumed linear removal: \( \frac{dPb}{dt} = F \cdot \alpha_k \cdot Ci \)

- \( Ci \) is the mixing ratio of tracer (e.g., 210Pb);
- \( F \) is the areal fraction that actually experiences precipitation;
- \( \alpha_k \) is the scavenging coefficient accounting for various scavenging processes.

The resultant scavenging 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.

Other related parameters:

- Cloud water content (CWC), having unit of cm⁻² water m⁻².
  - 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).

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° x 2.5° horizontal resolution and 47 vertical levels.
- Re-Pb-Be simulation option with Radon emission defined by Jacob et al. (1990).

210Pb Observations

- Latitudinal surface 210Pb distribution compiled by Preiss et al. [1996]
- RANDAB is a radionuclide database compiled from high-altitude aircraft and balloon measurements conducted during 1950s–1980s. It has specifically been used to evaluate simulated 210Pb in the upper troposphere and lower stratosphere (UTL/S).

NASA aircraft campaigns:

- PEM-West A, PEM-West B, TRACE-P, PEM-Tropics A, PEM-Tropics B, SUCCESS,
- POMEX, TSF, ICEollow, TC4

Model and Data

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Figure 1. Life cycle of 210Pb in the troposphere.

Parameterizations of precipitation scavenging

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The resultant scavenging 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. Parameters of the scavenging coefficient \( \alpha \)

Convecive Precipitation

<table>
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<tr>
<th>( \alpha_1 )</th>
<th>is determined by cloud formation rate and fixed condensation rate from cloud top to surface.</th>
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<tr>
<td>( \alpha_2 )</td>
<td>accounts for conversion from cloud to precipitation and cloud top precipitation.</td>
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Stratiform Precipitation

- \( \alpha_3 = \frac{\alpha_1 \cdot \alpha_2}{2} \)

- \( \alpha_3 \) is the overall BCS coefficient determined for an assumed 210Pb profile through calculations of the convective and stratiform fractions.

Re-evaporation

- 50% of aerosol is released back to ambient for the amount of precipitation evaporated.