Constraints From Airborne $^{210}\text{Pb}$ Observations on Aerosol Scavenging and Lifetime in GEOS-Chem

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**222Rn-210Pb simulation: sources and sinks**

**Scavenging processes:**
1. Scavenging in convective updrafts
2. Scavenging in stratiform (Large-Scale) cloud
3. Below-cloud scavenging (washout) by precipitation
4. Re-evaporation

**Focus of this work**

- Does the updated $^{222}\text{Rn}$ emission improve simulated $^{222}\text{Rn}$ and $^{210}\text{Pb}$?

- **What is the observation-constrained lifetime of $^{210}\text{Pb}$ aerosols?** How does it compare to previous work [H. Liu et al., JGR 2001; Q. Wang et al., ACP 2014]?

- What aspects of the scavenging parameterization need improving?
222Rn emission update and evaluation

- Incorporated recent 222Rn flux measurements in Europe, N. America, China, and Australia.
- We increased 222Rn fluxes over the SE China based on comparisons with 222Rn concentration and 210Pb dep flux.

Comparison with worldwide surface obs

Wet scavenging parameterization

- In convective updrafts: 
  \[ F = 1 - \exp \left( -a \cdot k \frac{\Delta Z}{\omega} \right) \]

- In stratiform clouds: 
  \[ F = a \cdot f \cdot [1 - \exp(-k \cdot \Delta t)] \]

- Below cloud (washout): 
  \[ F = f [1 - \exp(-k \cdot \Delta t)] \]

**Symbols:**

- \( F \): scavenging fraction
- \( \alpha \): fraction associated with cloud water, T-dependent look-up table
- \( k \): condensate-to-precip conversion rate; or scavenging rate constant in washout
- \( f \): cloud or precipitation areal fraction
- \( \Delta Z \): rising distance in updraft
- \( \omega \): updraft velocity

**Look-up table for \( \alpha \) in 3 temperature ranges \((^{210}\text{Pb} \text{ aerosol})\)**

<table>
<thead>
<tr>
<th>Temperature Range</th>
<th>cvrain</th>
<th>lsrain</th>
</tr>
</thead>
<tbody>
<tr>
<td>( T &lt; 237K )</td>
<td>1.0</td>
<td>1.0</td>
</tr>
<tr>
<td>( 237K \leq T &lt; 258K )</td>
<td>0.5</td>
<td>0.0</td>
</tr>
<tr>
<td>( T &gt; 258K )</td>
<td>1.0</td>
<td>1.0</td>
</tr>
</tbody>
</table>
$^{210}$Pb measurements from NASA aircraft campaigns provide strong constraint for scavenging parameterization

Table 1. List of NASA field campaigns where aircraft $^{210}$Pb and $^7$Be measurements were made by Jack Dibb (collaborator) and his colleagues at University of New Hampshire.

<table>
<thead>
<tr>
<th>Campaign</th>
<th>Region</th>
<th>Period</th>
</tr>
</thead>
<tbody>
<tr>
<td>PEM West A*</td>
<td>NW Pacific</td>
<td>September-October 1991</td>
</tr>
<tr>
<td>PEM West B*</td>
<td>NW Pacific</td>
<td>February-March 1994</td>
</tr>
<tr>
<td>PEM Tropics A*</td>
<td>southern tropical regions of the Pacific Ocean</td>
<td>August -September 1996</td>
</tr>
<tr>
<td>SUCCESS*</td>
<td>Atlantic Ocean, Gulf of Mexico, &amp; Eastern USA</td>
<td>April - May 1996</td>
</tr>
<tr>
<td>SONEX*</td>
<td>North Atlantic Flight Corridor</td>
<td>October-November 1997</td>
</tr>
<tr>
<td>PEM Tropics B</td>
<td>tropical Pacific</td>
<td>March-April 1999</td>
</tr>
<tr>
<td>TRACE P</td>
<td>NW Pacific</td>
<td>March-April 2001</td>
</tr>
<tr>
<td>TOPSE</td>
<td>North America</td>
<td>February - May 2000</td>
</tr>
<tr>
<td>INTEX NA</td>
<td>North America</td>
<td>summer 2004</td>
</tr>
<tr>
<td>INTEX B</td>
<td>North America</td>
<td>spring 2006</td>
</tr>
<tr>
<td>TC4</td>
<td>Eastern Pacific Ocean</td>
<td>July - August 2007</td>
</tr>
</tbody>
</table>

*These campaigns also include $^{10}$Be

- Calculated 5-day backward “trajectories” and sampled convective snow, convective rain, LS snow, and LS rain along the “trajectories”.
- Categorize observations according to precipitation types and amount.
Model experiments suggest that scavenging in LS mixed-phase clouds (237 K < T < 258 K) should be incorporated in the model.

Evidence is out there, e.g., riming in mixed-phase clouds (L. Qi et al., ACP 2017).
Evaluation w/ observed $^{210}$Pb dep. fluxes and surface conc.

**PREISS database:** Measurements of surface $^{210}$Pb conc. during 1960-1990.

- The updated emission resulted in higher deposition fluxes in the NH, but still reasonably consistent with observations.
- Model w/ the updated emission overestimates surface concentrations, suggesting stronger scavenging is required in the model.
- Increasing scavenging in large-scale precipitation improved simulated surface concentrations.
Evaluation w/ observed $^{210}$Pb seasonality

- Updated emission resulted in improved seasonality over NA and at high-latitude sites.
- Improved surface $^{210}$Pb concentrations with increased LS in-cloud scavenging.

**Observations**
- Updated Rn emission
- Standard
- Updated Rn emission + LS mixed-phase scav
- Updated Rn emission + LS mixed-phase scav + reduced LWC
Simulated tropospheric $^{210}$Pb lifetime

$$\text{lifetime} = \frac{\text{burden}}{\text{"wet + dry" deposition}}$$

<table>
<thead>
<tr>
<th>Lifetime (days)</th>
<th>Global Troposphere</th>
<th>60N - 90N</th>
<th>30N - 60N</th>
<th>15S - 15N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Standard</td>
<td>8.5</td>
<td>13.8</td>
<td>8.1</td>
<td>5.6</td>
</tr>
<tr>
<td>Emission update</td>
<td>8.3</td>
<td>12.3</td>
<td>8.1</td>
<td>5.5</td>
</tr>
<tr>
<td>lsrain100510 + cwc0.8</td>
<td>6.9</td>
<td>8.1</td>
<td>6.3</td>
<td>5.0</td>
</tr>
<tr>
<td>Israin101010</td>
<td>7.0</td>
<td>7.7</td>
<td>6.4</td>
<td>5.1</td>
</tr>
</tbody>
</table>

- Global tropospheric $^{210}$Pb lifetime reduced from 8.5 to ~ 6.9 days with LS mixed-phase cloud scavenging.
- Lifetime in northern high-latitude regions reduced ~ 42% (from 13.8 to 8.1 days), which contributed to the most change in global lifetime.
Summary and Conclusions

- Updated $^{222}$Rn emission improves model simulations of both $^{222}$Rn (conc. & seasonality) and $^{210}$Pb (seasonality).

- While the previously implemented ice/snow scavenging improves $^{210}$Pb simulation, our evaluation with aircraft/surface observations supports the incorporation of scavenging in mixed-phase clouds.

- **Our best estimated global $^{210}$Pb lifetime is $\sim 6.9$ days, which is $\sim 2$ days shorter than previously reported [H. Liu et al., JGR 2001].**

- The optimized scavenging parameters in this work may be only suitable for the MERRA input meteorology, but the resulting observation-constrained $^{210}$Pb aerosol lifetime can serve as a reference for other global models.

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