Interpreting Aerosol Lifetimes Using the GEOS-Chem Model and Constraints from Radionuclide Measurements

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Summary
- Aerosol lifetimes are indicative of scavenging efficiency, which remains poorly constrained in global models (Textor et al., 2006; Bond et al., 2013).
- Recently, Kristiansen et al. (2012) suggested (based on constraints provided by radionuclide measurements after the Fukushima Dai-Ichi nuclear power plant accident) that typical global mean aerosol lifetimes in present-generation global models (3-7 days) might be an under-estimate by a factor of two.
- We have developed the capability of GEOS-Chem to simulate the Fukushima Dai-Ichi nuclear accident. Simulated e-folding times agree within 20% of the measurements (site-mean) and differ by a factor of two only at certain tropical and high latitude sites.
- We show that differences between global mean aerosol lifetimes (which are controlled by initial fast removal for emissions into the boundary layer) and radionuclide e-folding times (which more closely reflect free tropospheric lifetimes) are about one order of magnitude.
- This study highlights the importance of careful interpretation of measurement-based constraints on aerosol lifetimes (Croft et al., in press).

1. Introduction
- Following the March 11, 2011 earthquake near Japan, a nuclear power plant accident at Fukushima Dai-Ichi caused massive radionuclide emissions.
- Examination of the subsequent decrease in 137Cs activity concentrations provides a constraint on global aerosol removal rates since 137Cs fully partitions to the aerosol phase immediately after emission.

GEOS-Chem Simulated 137Cs Column Burden on March 31, 2011

2. Lifetime Definitions

Mean Lifetime: \[ \tau_{6mo} = \frac{\sum_{i=0}^{6mo} C_i}{\sum_{i=0}^{6mo} S_i} \]

E-folding Time: \[ \tau_e = \frac{e^{-\ln(2)}}{S_3} \]

Instantaneous Lifetime: \[ \tau_{\text{instant}} = \frac{S_3 - S_2}{(C_3 - C_2)} \]

3. Regionally Varying Bias of Simulated 137Cs Surface Layer E-folding Times Relative to Measurements (in circles)

- Measurement e-folding times are from Kristiansen et al. (2012) and are shown in circles. GEOS-Chem simulation uses the radionuclide emissions data set of Stohl et al. (2012). 133Xe is treated as a passive tracer and 137Cs removal is similar that for sulfate.
- E-folding times are fit between days 20 and 80 after the March 11, 2011 earthquake.
- Regions of longer e-folding times at high latitudes are not in agreement with measurements suggesting possible errors in mixed-phase and ice cloud scavenging.
- The site-mean measurement e-folding time is 13.9 days. The simulated site-mean e-folding time is in good agreement (16.7 days).

4. Global Mean Lifetime is Strongly Dependent on Initial Quick Removal Before Aerosols Leave the Boundary Layer, E-folding Times are Longer and Reflect Aerosol Dynamics in Free Troposphere

- Sensitivity Simulations:
  - USFC: Same emissions as CTL spread uniformly in Northern Hemisphere surface layer.
  - U5K: Same as USFC, but all emissions in model layer at 5 km.
  - U7K: Same as USFC, but all emissions in model layer at 7 km.

5. Instantaneous Lifetimes in First Days After Emissions

Control Global Mean Aerosol Lifetimes

- Emissions into the boundary layer (CTL and USFC) have short instantaneous lifetimes in the first few days. This yields shorter global mean lifetimes (shown in Box 4) than for emissions outside the boundary layer (simulations U5K and U7K).
- After about 3 weeks, the radionuclides have reached a quasi-steady-state geographic distribution of concentrations. These lifetimes are then similar for all scenarios.

6. Aerosol Lifetimes Vary with Geographic Location and Altitude Reflecting the Differences in Scavenging Efficiency Between the Boundary Layer, Free Troposphere and Different Cloud Types

- Below 2 km, simulated zonal mean aerosol lifetimes are generally less than 7 days.
- There is a two-order-of-magnitude increase in these simulated mean lifetimes with respect to removal rates depending on altitude, particularly at high latitudes.
- Aerosol species show a simulated lifetime distribution similar to 137Cs.

References:

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