In-plume reduction and its implementation in the GEOS-Chem Hg simulation

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Date last modified: 13 May 2013

Observations collected at ground-based sites 7-15 km downwind of coal fired power plants (CFPPs) in the southeastern United States showed that the Hg(II) fraction measured in the atmosphere was a factor of ~2-4 lower than the Hg(II) fraction measured in CFPPs stacks (Edgerton et al., 2006). Similarly, measurements downwind of power plants by Weiss-Penzias et al. (2011) suggest that the average RGM fraction (21% and 8% at two different sites) is much lower than estimated by the local NEI 2002 emission inventory (~58 %). From airborne measurements downwind of a CFPP, ter Schure et al. (2011) suggest that the discrepancy does not reflect errors in the emission inventories but instead rapid in-plume reduction operating by a mechanism not found in the background atmosphere.

Several model studies also provide support for in-plume reduction of Hg(II) emitted from power plants (Seigneur et al., 2003; Lohman et al., 2006; Seigneur et al., 2006; Vijayaraghavan et al., 2008; Kos et al., 2011). Vijayaraghavan et al. (2008) incorporated this rapid reduction into the CMAQ regional Hg model with an explicit treatment of stack plume evolution. They found that this improved model performance for wet deposition in the Northeast U.S. Kos et al. (2011) implemented a 90:8:2 Hg(0):$Hg(II)_{gas}$:$Hg(II)_{particle}$ speciation in the GRAHM global Hg model to better match surface concentrations of RGM and PBM over North America.

In order to consider this process in GEOS-Chem, we modified the CFPP Hg emission partitioning from the original 56.8% Hg(0), 39.6% $Hg(II)_{gas}$, 3.6% $Hg(II)_{particle}$ in the NEI and NPRI inventories to 86.5% Hg(0), 9.9% $Hg(II)_{gas}$, 3.6% $Hg(II)_{particle}$ over the US and Canada. This effectively assumes that 75% of the CFPPs Hg(II) emissions are reduced to Hg(0) in the immediate vicinity of power plants, consistent with Edgerton et al. (2006) and Weiss-Penzias et al. (2011). More details are available in Zhang et al. (2012).

In v9-01-02, the Hg(0):$Hg(II)_{gas}$:$Hg(II)_{particle}$ speciation profile of emissions from fossil fuel combustion in the GEIA 2005 global anthropogenic emission inventory (Pacyna et al., 2010) are modified from 50:40:10 to 86.5:9.9:3.6 (Amos et al., 2012). The change in speciation is based on Zhang et al. (2012). This modification is made for all grid cells containing anthropogenic emissions.

In v9-02, a logical `LInPlume` is added in `mercury_mod.F` to control the in-plume reduction. If enabled, the coal fired power plants emission speciation will be modified. In the US and Canada, the location and emission rate from CFPPs are available in the NEI2005 and NPRI2005 emission inventories, respectively. So only the grid points with non-zero CFPP emissions are modified; based on its strength of CFPP Hg emissions. Outside of US and Canada, we modify anthropogenic emissions based on the overall fraction of total anthropogenic emissions from fossil fuel combustion as described by Amos et al. (2012).

This implicit inclusion of Hg(II) in-plume reduction in the model comes with the important caveats that a chemical mechanism has not been identified (Lohman et al., 2006) and that there are significant uncertainties associated with both the speciation of anthropogenic emission inventories (AMAP/UNEP, 2008) and the methods for measuring atmospheric Hg (Gustin and Jaffe, 2010).

Streets et al. (2011) present historical anthropogenic emissions for continental-scale regions at decadal time resolution since the year 1850. For each decade, we separate out the fraction of total anthropogenic emissions from fossil fuels and scale global Hg(0), $Hg(II)_{gas,}$ and $Hg(II)_{particle}$_emissions so that fossil fuel emissions follow the 86.5:9.9:3.6 ratio in Amos et al. (2012). Contact Bess Corbitt (sturges@fas.harvard.edu) if you are interested in using the scaled historical anthropogenic emissions before they are implemented in the standard code.

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