Text S1. Discussion of Bromine Chemistry

The current version of HYSPLIT-Hg does not include the potential oxidation of Hg(0) by bromine (Br/BrO), in part because of uncertainty in estimating the concentrations of reactive bromine in the atmosphere (De Simone et al., 2014; Saiz-Lopez and von Glasow, 2012; Simpson et al., 2015). Bromine is used as the primary oxidant in GEOS-Chem-Hg (Br option) (e.g., Song et al., 2015) and in sensitivity studies with CTM-Hg (Seigneur and Lohman, 2008), ECHMERIT (De Simone et al., 2015), and WRF/Chem (Gencarelli et al., 2015). Bromine along with other oxidants (e.g., OH• and O3) have been used in other analyses, including GEOS-Chem (Kikuchi et al., 2013), CMAQ-Hg (Zhu et al., 2015), CAM-Chem-Hg (Lei et al., 2013, as a sensitivity analysis), and a recent version of GRAHM (Kos et al., 2013). Many recent atmospheric mercury model analyses, like the present analysis, do not include bromine-mediated oxidation. Examples include GEOS-Chem (O3-OH option) (e.g., Weiss-Penzias et al., 2015), CMAQ-Hg (Baker and Bash, 2012; Bieser et al., 2014; Holloway et al., 2012; Lin et al., 2012; Myers et al., 2013; Zhu et al., 2015), CTM-Hg (base version) (Lohman et al., 2008; Seigneur et al., 2006; Seigneur et al., 2004) , DEHM (Christensen et al., 2004; Hansen et al., 2015), ECHMERIT (base version) (De Simone et al., 2015; De Simone et al., 2014), GLEMOS (non-polar regions) (Bieser et al., 2014), MSCE-HM (Travnikov, 2005), REMSAD (e.g., Bullock et al., 2008), TEAM (Bullock et al., 2008), and WRF/Chem (base version) (Gencarelli et al., 2014; Gencarelli et al., 2015). Whatever the chemical mechanism used, most atmospheric mercury models have relatively similar, overall, net Hg(0) oxidation rates, as they are constrained by the net emissions of Hg(0) and the measured concentrations of Hg(0) in the atmosphere.

Some studies have found that the use of bromine produced similar results to using O3/OH• as oxidants (De Simone et al., 2015; Holmes et al., 2010). Gencarelli et al. (2015) found an improvement in model estimated wet deposition results using a Br/BrO oxidation mechanism, but the overestimated wet deposition found with the O3/OH• mechanism was produced using the (relatively fast) nominal reaction rates discussed above. As will be seen below, even with the relatively “slow” rates for the O3 and OH• oxidation reactions used in this work, the HYSPLIT-Hg model tends to produce ground-level concentrations of non-Hg(0) mercury forms (Hg(II) , Hg(p), and Hg2s) equal to or greater than those measured.

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