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Surface reactions can alter both perceived and actual composition of atmospheric Hg(II)

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Mercury is a persistent and bioaccumulative environmental pollutant, entering the atmosphere mostly in elemental form and leaving in a variety of oxidized forms. However, current detection methods distinguish only three operationally defined forms: elemental, gaseous oxidized, and particulate mercury. The knowledge of molecular speciation is limited and comes mostly from computational studies, hindering our understanding of atmospheric mercury chemistry and global cycling. Achieving molecular speciation of oxidized mercury (Hg(II)) in atmospheric measurements is a major challenge because of its extremely low abundance, requiring pre-concentration on adsorbents. There is a concern that the speciation of adsorbed Hg(II) can be altered in exchange reactions with itself and with co-adsorbed atmospheric chemicals. Here we present the results of our experimental investigation of gas-surface reactions and exchange reactions involving Hg(II) and several other chemical species. These reactions were studied in aqueous solutions and on surfaces, and the products were analyzed at a molecular level in gaseous, dissolved, and solid forms. Molecular-level detection was made possible by using chemical ionization and electrospray ionization mass spectrometries, which provided sufficiently high sensitivity for direct detection of Hg(II) in laboratory experiments, avoiding the pre-concentration step. In all cases, we observed binding of gaseous Hg(II) to surfaces, followed by rapid formation of exchange products, some of which could be readily volatilized back into gas phase. We propose that a similar exchange may occur both on the surfaces of aerosols in the atmosphere and on adsorbents during sample collection, scrambling the actual and perceived speciation of atmospheric Hg(II).