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High levels of reactive gaseous mercury observed at a high elevation research laboratory in the Rocky Mountains

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Abstract. The chemical cycling and spatiotemporal distribution of mercury in the troposphere is poorly understood. We measured gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and particulate mercury (Hg_p) along with carbon monoxide (CO), ozone (O_3), aerosols, and meteorological variables at Storm Peak Laboratory at an elevation of 3200 m a.s.l., in Colorado, from 28 April to 1 July 2008. The mean mercury concentrations were 1.6 ng m^{-3} (GEM), 20 pg m^{-3} (RGM) and 9 pg m^{-3} (Hg_p). We observed eight events of strongly enhanced atmospheric RGM levels with maximum concentrations up to 137 pg m^{-3} . RGM enhancement events lasted for long time periods of 2 to 6 days showing both enriched level during daytime and nighttime when other tracers (e.g., aerosols) showed different representations of boundary layer air and free tropospheric air. During seven of these events, RGM was inversely correlated to GEM (RGM/GEM regression slope ~ -0.1), but did not exhibit correlations with ozone, carbon monoxide, or aerosol concentrations. Relative humidity was the dominant factor affecting RGM levels with high RGM levels always present whenever relative humidity was below 40 to 50%. We conclude that RGM enhancements observed at Storm Peak Laboratory were not induced by pollution events and were related to oxidation of tropospheric GEM. High RGM levels were not limited to upper tropospheric or stratospherically influenced air masses, indicating that entrainment processes and deep vertical mixing of free tropospheric air enriched in RGM may lead to high RGM levels throughout the troposphere and into the boundary layer over the Western United States. Based on backtrajectory analysis and a lack of mass balance between RGM and GEM, atmospheric production of RGM may also have occurred in some distance allowing for scavenging and/or deposition of RGM prior to reaching the laboratory. Our observations provide evidence that the tropospheric pool of mercury is frequently enriched in divalent mercury, that high RGM levels are not limited to upper tropospheric air masses, but that the build-up of high RGM in the troposphere is limited to the presence of dry air.

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