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Estimating surface fluxes of very short-lived halogens from aircraft measurements over the tropical Western Pacific

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We use an inverse model approach to quantitatively understand the ocean flux and atmospheric transport of very short-lived halogenated species (VSLS) measured during the coordinated NERC CAST and NCAR CONTRAST aircraft campaigns over the Western Pacific during January/February 2014. To achieve this we have developed a nested GEOS-Chem chemistry transport model simulation of bromoform (CHBr3) and dibromomethane (CH2Br2), which has a spatial resolution of 0.25° (latitude) × 0.3125° (longitude) over the tropical Western Pacific region, and fed by boundary conditions from a coarser version of the model. We use archived 3-hourly 3-D fields of OH and j-values for CHBr3 photolysis, allowing us to linearly decompose these gases into tagged contributions from different geographical regions. Using these tagged tracers, we are able to use the maximum a posteriori probability (MAP) approach to estimate the VSLS sources by fitting the model to observations. We find that the resulting VSLS fluxes are significantly different from some previous studies. To interpret the results, we describe several observation system simulation experiments to understand the sensitivity of these flux estimates to observation errors as well as to the uncertainty in the boundary condition imposed around the nested grid.