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Global oceanic emissions of very short-lived (VSL) halocarbons in a chemistry-climate model

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The global CAM-Chem chemistry-climate model contains parameterisations of stratospheric and tropospheric chemistry. Its scope has been extended to include halogen chemistry in the troposphere. This comprises natural sources of very short-lived (VSL) halocarbons from the oceans; reactive chlorine, bromine and iodine species; related photochemical, gas-phase and heterogeneous reactions, as well as wet and dry deposition for relevant species. We have derived an emission inventory for VSL bromocarbons ($CHBr_3$, CH_2Br_2, CH_2BrCl, $CHBrCl_2$, $CHBr_2Cl$) and iodocarbons (CH_2ICl, CH_2IBr, CH_2I_2) using a compilation of aircraft campaigns and more sparse observations in the marine boundary layer (MBL), respectively. Emissions for methyl iodide (CH_3I) in CAM-Chem are based on the inventory from a previous modelling study while the longer lived methyl bromide (CH_3Br) and organochlorides are set as lower boundary conditions.

Reported correlations between the abundance of bromocarbons and areas of high primary productivity (as indicated by SeaWiFS satellite data for chlorophyll-a concentration) have been used to constrain the emission fields for most VSL halocarbons over the tropical oceans (defined as 20° S - 20° N). These emission sources have been extended to the mid- and high-latitude oceans, where we consider latitudinal variation in addition to 2.5 higher emission fluxes over the coastal areas to account for the stronger sources there compared to the open ocean. Observed concentration ratios among bromoform ($CHBr_3$) and other bromocarbons as well as subsequent comparisons of modelled bromocarbon mixing ratios with a composite of aircraft observations have been exploited to derive their total emission fluxes. Overall, these are in good agreement with emission magnitudes determined by previous model studies. The simulated vertical profiles of bromocarbons and methyl iodide, which have atmospheric lifetimes ($\sim$ 7 to 150 days) long enough for them to be transported to the upper troposphere within deep convection areas, are similar in magnitude and vertical distribution to airborne observations in the troposphere. The total emission fluxes of the remaining iodocarbons (CH_2ICl, CH_2IBr, CH_2I_2) and molecular iodine (I_2), with shorter lifetimes on the order of a few hours to seconds and therefore more relevant as sources of reactive halogens in the MBL, have been determined following previous reports from the scientific literature and comparisons with available observations in the MBL.

Further work will quantify the potential of bromine and iodine chemistry to influence the oxidation capacity of the atmosphere.