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Online calculation of global marine halocarbon emissions in the chemistry climate model EMAC

Sinikka T. Lennartz (1), Gisèle Krysztofiak-Tong (2,3), Björn-Martin Sinnhuber (3), Christa A. Marandino (1), Susann Tegtmeier (1), Kirstin Krüger (4), Franziska Ziska (1), and Birgit Quack (1)

(1) GEOMAR Helmholtz-Zentrum für Ozeanforschung Kiel, Kiel, Germany (slennartz@geomar.de), (2) LPC2E, UMR7328, CNRS-University of Orléans, Orléans, France (gisele.krysztofiak@cnrs-orleans.fr), (3) Karlsruhe Institut of Technology, Institut for Meteorology and Climate Research, Karlsruhe, Germany (bjoern-martin.sinnhuber@kit.edu), (4) University of Oslo, Department of Geosciences, Oslo, Norway (kirstin.krueger@geo.uio.no)

Marine produced trace gases such as dibromomethane (CH2Br2), bromoform (CHBr3) and methyl iodide (CH3I) significantly impact tropospheric and stratospheric chemistry. Marine emissions are the dominant source of halocarbons to the atmosphere, and therefore, it is crucial to represent them accurately in order to model their impact on atmospheric chemistry. Chemistry climate models are a frequently used tool for quantifying the influence of halocarbons on ozone depletion. In these model simulations, marine emissions of halocarbons have mainly been prescribed from established emission climatologies, thus neglecting the interaction with the actual state of the atmosphere in the model. Here, we calculate halocarbon marine emissions for the first time online by coupling the submodel AIRSEA to the chemical climate model EMAC. Our method combines prescribed water concentrations and varying atmospheric concentrations derived from the model instead of using fixed emission climatologies. This method has a number of conceptual and practical advantages, as the modelled emissions can respond consistently to changes in temperature, wind speed, possible sea ice cover and atmospheric concentration in the model. Differences between the climatology-based and the new approach (2-18%) result from consideration of the actual, time-varying state of the atmosphere and the consideration of air-side transfer velocities. Extensive comparison to observations from aircraft, ships and ground stations reveal that interactively computing the air-sea flux from prescribed water concentrations leads to equally or more accurate atmospheric concentrations in the model compared to using constant emission climatologies. The effect of considering the actual state of the atmosphere is largest for gases with concentrations close to equilibrium in the surface ocean, such as CH2Br2. Halocarbons with comparably long atmospheric lifetimes, e.g. CH2Br2, are reflected more accurately in EMAC when compared to time series of ground based stations than shorter lived gases, suggesting that their atmospheric seasonality is driven by seasonality in transport rather than in emissions. Results of this study further indicate that the accurate presentation of shorter lived gases such as CH3I in models requires seasonally resolved water concentration climatologies.