

## Interaction and variability of very short-lived substances from the tropical Indian Ocean to the stratosphere

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Halogenated very short-lived substances (VSLSs) are naturally produced in the ocean and emitted to the atmosphere. When transported to the stratosphere, these compounds can have a significant influence on the ozone layer and climate. During a research cruise on RV Sonne in the subtropical and tropical west Indian Ocean (WIO) in July and August 2014, we measured the VSLSs, methyl iodide(CH3I, 3.5 days atmospheric lifetime), bromoform (CHBr, 17 days) and dibromomethane (CH2Br2, 150 days), in surface seawater and the marine atmosphere to derive their emission strengths. We found that the WIO is a strong source for CHBr3 (910 pmol m-2 h-1), very strong source for CH2Br2 (930 pmol m-2 h-1) and an average source for CH3I (460 pmol m-2 h-1). We derive the strongest VSLS emissions south of Madagascar and in the trade wind regime from 5 to 10 S above the open ocean upwelling region of the Seychelles-Chagos-thermocline ridge, where we also observed enhanced phytoplankton growth.

Using a Lagrangian transport model, we calculated the direct contribution of observed VSLS emissions to the stratospheric halogen burden during the Asian summer monsoon. Furthermore, we investigated the interannual variability of VSLS transport from a larger area of the WIO surface to the stratosphere for 2000–2015.

The stratospheric entrainment of VSLS tracers from the WIO shows a distinct annual cycle associated with the Asian monsoon, which experiences two main pathways. On very short timescales, especially relevant for the shortest-lived compound (CH3I), convection above the Indian Ocean lifts oceanic air masses and VSLSs towards the tropopause. On a longer timescale, the Asian summer monsoon circulation transports longer-lived VSLSs (CHBr3 and CH2Br2) towards India and the Bay of Bengal, where they are lifted with the monsoon convection and reach stratospheric levels in the south-eastern part of the Asian monsoon anticyclone. Over the 16 year time series, a slight increase in entrainment from the WIO to the stratosphere is found for all VSLS tracers and during all seasons. The interannual variability shows a relationship with sea surface temperatures in the WIO as well as the El Niño–Southern Oscillation (ENSO). During boreal spring of El Niño, enhanced stratospheric entrainment of VSLS from the tropical WIO is caused by positive sea surface temperature anomalies and enhanced vertical uplift above the WIO. During boreal fall of La Niña, stronger entrainment is related to enhanced atmospheric upward motion over the East Indian Ocean and a prolonged Indian summer monsoon season. Overall, the tropical WIO is a significant source region for the halogen loading in the stratosphere with pronounced seasonality due to ocean and atmospheric transport variability associated with the Asian monsoon.