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Oceanic VSLS contribution to the stratosphere: Comparing tropical East Atlantic with tropical West Pacific emissions

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Halogenated very short-lived substances (VSLS) are expected to contribute significantly to the stratospheric halogen loading and therefore to the stratospheric ozone chemistry. Tropical waters and upwelling regions in the oceans have been identified as potentially important source regions for the naturally produced VSLS, such as volatile brominated and iodinated halocarbons. Our understanding of the transport of VSLS from the marine boundary surface into the stratosphere is crucial to estimate their contribution to stratospheric halogen loading. In particular, the chemical degradation, wet and dry deposition as well as their transport through the free troposphere up to the stratosphere play important roles in determining whether VSLS and their organic product gases are able to reach the stratosphere.

In this study we investigate the VSLS emissions in the tropical East Atlantic and tropical West Pacific and their contribution to the stratospheric halogen loading. For this purpose we use the Lagrangian particle dispersion model FLEXPART, which simulates transport, small scale mixing, washout and photochemical decay of three VSLS (bromoform, dibromomethane and methyliodide). The transport simulations are based on the VSLS sea-to-air flux obtained from the tropical East Atlantic in May/June 2010 and from the tropical Western Pacific in October 2009. While relatively large brominated VSLS fluxes are found in the Mauritanian upwelling in the East Atlantic, the VSLS transport is most efficient in the Western Pacific atmosphere, a region characterized by a high convective activity throughout the year. We show, based on the evaluations for the two tropical campaigns that the peak emissions of brominated VSLS together with strong convective transport lead to significant abundances in the TTL, which are comparable to available upper air measurements. For methyliodide however the Western Pacific emission-based profiles are larger than observations, which suggests that existing measurements may not be representative and methyliodide could have a larger impact on the stratospheric halogen loading than assumed so far. Finally, we will relate the results of this paper also to past ship campaigns in the tropical Atlantic and to the recent SHIVA Sonne expedition to the South-China Sea during November 2011.