Trace gas emissions from tropical oceans influence the stratospheric ozone layer

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The halogens chlorine, bromine and iodine are highly efficient at destroying atmospheric ozone. Rising concentrations of these compounds from human activities have led to depletion of global stratospheric ozone over the last decades, and formation of the Antarctic "ozone hole". Whereas the chlorine supply is dominated by anthropogenic emissions, a major part of the bromine and iodine is supplied by short-lived organic trace gases with oceanic sources, entering the stratosphere principally in the tropics. The Western Pacific is a projected hot spot area for both oceanic sources and stratospheric entrance region of the trace gases. In order to reduce uncertainties in the amount of naturally emitted halogen-containing compounds reaching the stratosphere and the resulting ozone depletion, a large campaign was performed in the western Pacific, combining measurements from the German research vessel SONNE, the German research aircraft DLR-FALCON as well as land and space-based platforms.

Since the 1970s it is known that gases containing chlorine and bromine emitted by human activities deplete stratospheric ozone. 200 nations committed themselves to limit and abandon the emissions of the industrially produced chlorofluorocarbons (CFC) in the Montreal Protocol in 1989. Their goal to protect the ozone layer, close the existing ozone hole and therewith decrease UV radiation at the surface is underway, although the ozone hole and durable CFCs still exist in the stratosphere. In concert with climate change the future development of the ozone layer is very uncertain, not least because the contribution of natural halogenated compounds to the destruction of ozone is hardly known. Strong sources of brominated and iodinated organic compounds for the atmosphere are found in the oceans. Regionally enhanced phytoplankton and photochemical reactions in the open ocean and macro algae as well as anthropogenic sources in coastal regions are known contributors to the highly variable marine and atmospheric concentrations of substances such as bromoform ($CHBr_3$), dibromomethane (CH_2Br_2) and methyl iodide (CH_3I).

Their highly variable oceanic emissions cause major uncertainties in the contribution of the natural sources to stratospheric halogens and therewith ozone depletion. A good spatial and temporal resolution of emissions and the understanding of their variability are needed for an accurate climate-ozone-interaction modeling. This includes the resolution of gradients between coastal, near-shore and open ocean emissions, the revelation of the diversity of emissions in variable environments and their controls, as well as the magnitude of global air sea fluxes and the possible effects of climate change on the fluxes.

Figure 1: The research aircraft FALCON from the DLR measures and follows air masses that have been investigated on the SONNE (21. November 2011).

The tropical oceans have been identified as potentially important source regions for various halogenated trace gases, where marine emissions have the potential to enter the stratosphere due to rapid uplift of surface air to the tropical tropopause layer in form of deep convection. Especially the tropical western Pacific is a largely uncharacterized region for the oceanic compounds, where the research cruise SO218 of the German research vessel SONNE was conducted by the Chemical Oceanography in cooperation with the Marine Meteorology research unit. From 15 to 29 November 2011 an international crew of European, Malaysian and Philippine scientists investigated the South China and Sulu Sea within the frame work of the EU-

Figure 2: Local Malaysian research boats meet RV Sonne for the exchange of water and air samples off the coast of Borneo (19./21. November 2011).

project SHIVA (Stratospheric ozone: Halogen Impacts in a Varying Atmosphere; http://shiva.iup.uni-heidelberg.de).

The instruments on board made quasi-continuous measurements of a suite of trace gases in both seawater and air to determine actual sea to air fluxes. The atmospheric structure was determined by intense radio and ozone sounding and in situ and satellite measurements of phytoplankton groups informed about biogeochemical conditions during the ship expedition.

The research during the cruise was coordinated with research flights of the Falcon Aircraft (Figure 1) from the German Aerospace Center's (DLR) which was stationed in Miri on Borneo for this project. The air craft over passed the ship several times and its atmospheric sampling started at the same height as the ship measurements and followed the same air masses up to an altitude of 13 km. In order to obtain near-coastal samples, which were analyzed on board the SONNE for their trace gas and plankton content, meetings with coastal research cruises from our Malaysian partners were organized and successfully conducted in the open ocean (Figure 2),

First results show generally enhanced concentrations of brominated and iodinated trace gases in the water, being very high in some shallow shelf-regimes and close to the coasts (Figure 3), while the atmospheric abundances were generally rather low. Strong oceanic emissions with rapid dilution in the atmosphere indicate that the cruise region comprises major source areas of biogenic halogen compounds capable of damaging the stratospheric ozone layer. Measurements and models indicate that the vertical transport in large tropical thunderstorms is a very important factor for the transport of the ozone relevant gases into the upper tropical troposphere, from where these gases may reach the lowermost stratosphere.

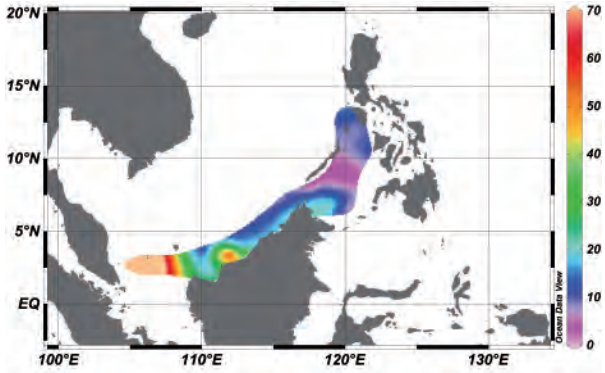

Figure 3: Concentrations (pmol/L) of Bromoform ($CHBr_3$) in surface water of the South China and Sulu Sea.

The results of the SHIVA-SONNE campaign about the natural and oceanic sources of ozone depleting substances will help to better predict the rate, timing and climate sensitivity of ozone-layer recovery in response to the decline of the industrial "ozone killers" like chlorofluorocarbons (CFC) which interact with the oceanic halogenated organic trace gases. Further spatial and temporal wide measurements of the biogenic compounds containing chlorine, bromine and iodine are needed in order to provide a profound surface forcing for simulations of the future development of the ozone layer incorporating these natural sources of halogens under the influence of anthropogenic change.