The SHIVA Western Pacific Campaign in Fall 2011

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ABSTRACT This paper briefly summarizes the activities and observations made during the SHIVA (Stratospheric Ozone: Halogen Impacts in a Varying Atmosphere) Western Pacific campaign in fall 2011, here referred to as the SHIVA South China Sea (SCS) campaign. The campaign addressed investigation of emissions of so-called halogenated very short-lived substances (VSLS) from the South China Sea (SCS), their atmospheric transport and transformation, their contribution to the budget of ozone destroying halogens in the stratosphere. Key findings of the SHIVA campaign are that, (a) the SCS is indeed a prominent emission region for halogenated VSLS, mostly due to the emissions of micro- and macro-algae, (b) in the boundary layer of the marine atmosphere typical VSLS mixing ratios were 3.6 – 13.3 ppt and in the upper troposphere 4.1±0.6 ppt, which may well explained a total source gas (SG) and product gas (PG) injection of [VSLS] = 4 – 5 ppt into the global stratosphere.

(**Keywords:** oceanic-atmospheric coupling, photochemistry, stratospheric ozone)

INTRODUCTION

It is now well known that certain ozone depleting substances (ODSs) are broken down by solar radiation in the stratosphere, releasing the halogen elements: chlorine, bromine and iodine. The halogens are highly efficient in destroying ozone in the stratosphere, and rising halogen concentrations from human activities have led to depletion of global stratospheric ozone over the last three decades, and formation of the Antarctic "ozone hole". ODSs enter the stratosphere principally in the tropics, where ascending warm air carries them aloft. SHIVA aims to reduce uncertainties in the amount of naturally emitted halogen-containing ODSs so called very short-lived substances (VSLS) reaching the stratosphere, and the resulting ozone depletion, in a climate that is changing now and it is likely to do so in future. Climate feedbacks between the emissions and

transport of ODSs exist, particularly in the tropics where even short-lived ODSs of natural origin (e.g., emitted by the oceans, and by marine and terrestrial organisms) can enter the stratosphere in powerful deep convective systems.

Along these lines, the project SHIVA (Stratospheric ozone: Halogen Impacts in a Varying Atmosphere) received funding within the European Union's 7th framework programme (e.g., see http://shiva.iup.uni-heidelberg.de/). In its core, SHIVA's objectives were:

- To study the oceanic emission strengths of a suite of halogenated source gases
- To investigate their atmospheric transport and transformation into the relevant product gases as a function of larger suite of oceanic (physical, chemical and biological) parameters and atmospheric, i.e., macro- and meso-scale weather patterns.
- To analyse the past, present and likely future trend of the total halogen burden in the stratosphere
- To assess and predict the impact of long and short-lived halogenated source gases (SG) and their inorganic product gases (PG) for past, present and future ozone within the upper troposphere, TTL and global stratosphere

Since ODS emissions are known to be strong in tropical oceans and their margins, and convective transport is known to be most intense in the Western Pacific region, a field campaign was performed in the South China Sea in fall (Sept. to Dec.) 2011 together with several Malaysian partners. It comprised investigations in the laboratory, at different ground-based stations, from local research ships, the research vessel (RV) Sonne, the DLR (German aerospace center) aircraft Falcon, and space-based platforms. The present paper briefly summarizes these activities and gives an overview on research results achieved to date.

SPECIFIC OBJECTIVES AND AIMS OF THE SHIVA SCS CAMPAIGN

Among a wider range of SHIVA objectives, specific objectives of the SHIVA SCS field campaign were to investigate:

- The production and emissions of VSLS from a large range of different marine environment and biota in water and air were studied in their natural environments as well as in the laboratory, e.g., different samples of marine biota were taken by local boats and from the shore and later cultivated in the laboratory.
- The biological productivity in the ocean water per se, e.g., investigated by local boat, research vessels and satellite-based instruments.
- The VSLS emissions in water and air of coastal zones having different (macro-) biota, such as mangrove forests, meadows of seaweed and sea-grass, algae plumes, corals, and others, e.g., investigation with instruments deployed on local boats in the intertidal zone, and in the laboratory.
- In the waters of the open ocean and the overlying atmosphere meso-scale VSLS emission pattern, e.g. by investigation using a large research vessel, and aircraft-based instrumentation.
- The transport of VSLS loaded air masses into the free and upper troposphere, e.g. by aircraft-based investigations.
- The transformation of VSLS into their inorganic decay products within the marine boundary layer and the troposphere, e.g., by ground, research vessel, aircraft, and satellite-based instrumentation.

Dedicated field observations however dedicated also need a good deal of guidance, and forecasts inferred from a larger range of existing theoretical tools and models. Therefore a suite of models were used such as weather assimilation and forecast models, process models addressing the regional meteorology, in particular meso-scale convective models, air mass trajectory models and global chemistry transport and climate models.

In fall 2011, the SHIVA SCS field campaign was performed along the coastlines of Peninsular Malaysia and West Malaysia in Borneo (Figure 1). It involved (a) land-based investigation teams working at Tawau and Semporna, (b) a cruise of the Research Vessel (RV) Sonne leading from Singapore to Manila (Nov. 15 - 29, 2011 (Figure 2), (c) local boat cruises departing from Port Dickson (2°31′N, 101°48′E) on Oct. 5, 2011, from Kuching (1°33′36″N, 110°20′42″E) on Nov. 11, 2011, Kota Kinabalu (5°58′17″N, 116°05′43″E) on Nov. 23, 2011,and Semporna (4°29′0″N 118°37′0″E) on Nov. 26, and 27, 2011 and (d) deployment of the DLR Falcon aircraft with 16 sorties undertaken from Miri (Nov. 12 – Dec. 12, 2011) (Figure 3).

On its cruise from Singapore to Manila, the crew on the RV Sonne studied the marine production and emission of VSLS as a function of physical, chemical and biological parameters in deep and up to near surface waters and the ambient atmosphere. In addition, the German DLR

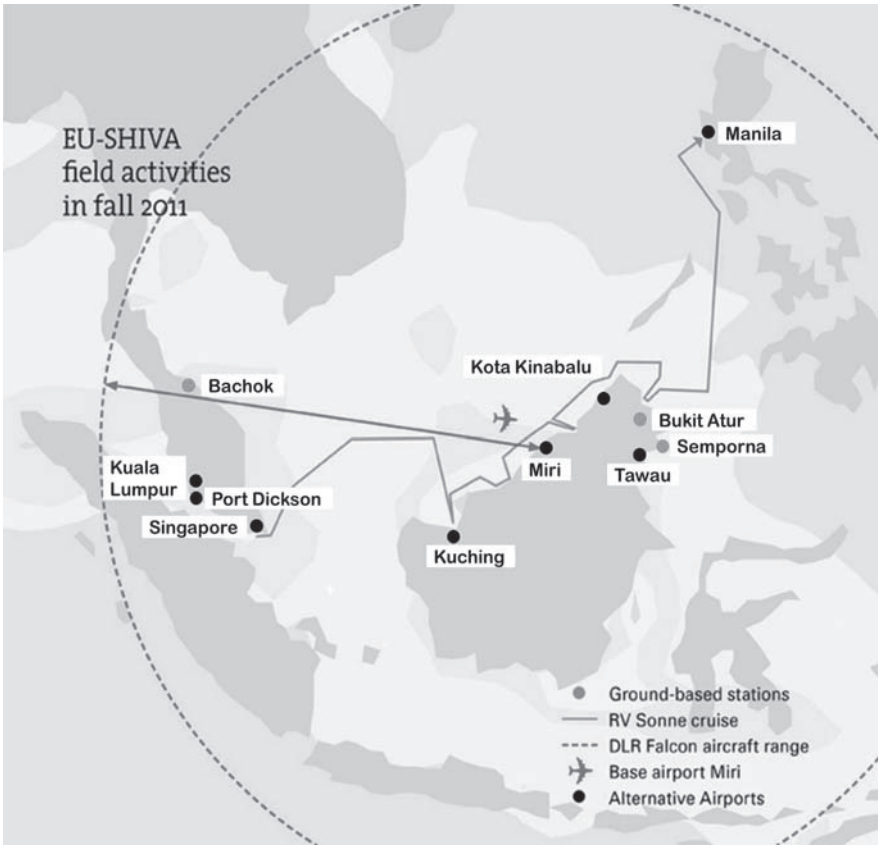

Figure 1. Schematics of the SHIVA Western Campaign activities in fall 2011

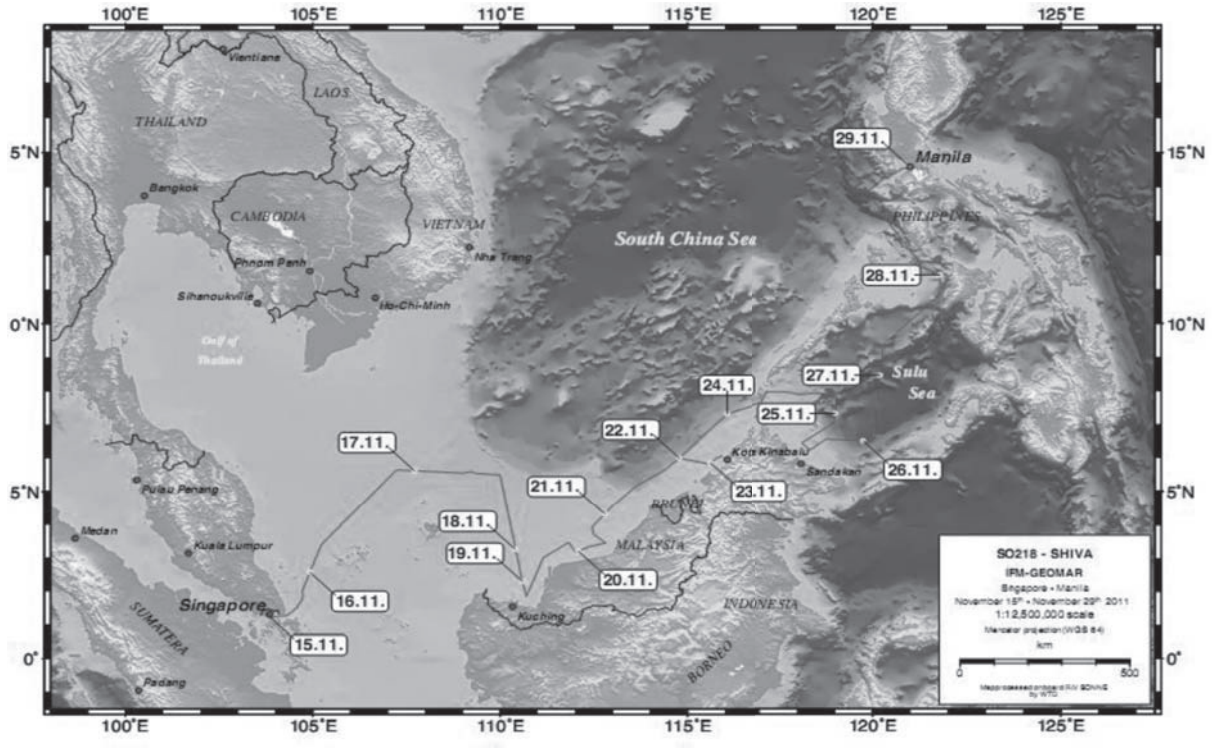

Figure 2. The RV Sonne ship track leading from Singapore to Manila between Nov. 15 and 29, 2011.

Falcon aircraft performed a series of sorties from Miri in Sarawak to study the emissions, atmospheric transport and transformation of VSLS around Borneo and the South China Sea (Figure 3).

Additional radio and ozone sondes were also launched from land by UM and from the RV Sonne by IFM-GEOMAR, in order to support the post campaign meteorological analysis and modelling activities.

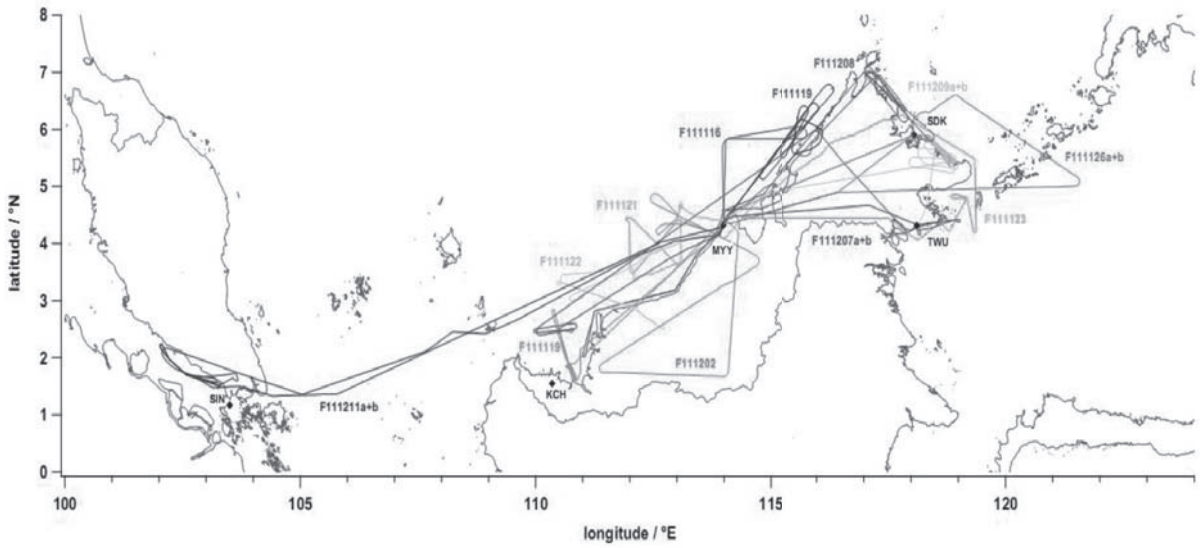

Figure 3. Overview on DLR Falcon flight patterns for sorties from Miri (Sarawak) between Nov. 10 and Dec. 12, 2011.

Satellite observations by MERIS and ENVISAT were used to watch the biological activity of the studied water via ocean colour measurements (AWI) and the atmospheric abundance of some crucial halogen oxides (BrO, and IO) was monitored (BIRA-IASB and UBREM). Furthermore, teams from the AWI, DLR, UM, ULEEDS and MMD provided the meteorological forecasting and analysis contributed by supportive modelling studies to the field work.

RESULTS

Even though the retrieval and interpretation of the data collected during the SHIVA SCS campaign is still ongoing, the following results have emerged:

- The largest VSLS emissions from marine biota ever recorded were observed from the laboratory and field studies, notably from red algal species, with wide variations both between and within species. The emission of VSLS also appeared to be light dependent, and differences were seen between measurements made in the laboratory and in the field. Significant emissions from intertidal sediments along open coasts and within mangroves were observed.
- The deep and surface waters probed in the South China Sea and Sulu Sea during the RV Sonne cruise, as well as during the local boat cruises near the coasts of Kuching, and Kota Kinabalu have been identified as strong sources for some of the halogenated VSLS. In particular, oceanic water enhancements of $CHBr_3$ and CH_2Br_2 tended to correlate with haptophyte, chrysophyte, prasinophyte, and *Prochlorococcus* concentrations, while CH_{3I} enhancements seemed to correlate with the cyanobacteria abundance. At the same time when high to very high concentrations of VSLS were found, the atmospheric VSLS concentrations were comparatively low. Both findings are indicative of large water to air emissions of VSLS from the open ocean and coastal waters of the SCS and Sulu Sea into the atmosphere.
- During the sorties of the DLR Falcon aircraft from Miri a variety of atmospheric processes were studied which provide novel information on the transport and transformation of halogenated VSLS in the troposphere and their potential contribution of VSLS to the stratospheric halogen budget.

These observations included the identification of major VSLS emission regions, i.e. around Tawau, Semporna and Sandakan and the Straits of Malacca and to a lesser extend along the northwestern coast of Borneo, and a sign of uplift of VSLS by shallow and deep convection. Overall in the marine boundary layer, total VSLS bromine was in the range of 3.9 – 10.7 ppt, with a mean value of 5.7 ppt, which compares well to 3.6 – 13.3 ppt, with a mean value of 8.4 ppt reported by WMO (2011).Above 11 km (3.72 ± 0.60 ppt) ppt of VSLS bromine was found as compared to 3.5 ppt reported by WMO (2011). The latter observation can be interpreted by fast convective uplift of near surface air into the upper troposphere frequently occurring in the SCS, a statement for which evidence was also found in the concentration profile of atmospheric trace gas species (e.g., CO, O_3, and CH_2O). Signatures of other relevant trace gases, tentatively emitted by natural biogenic emissions, biomass burning-related activities as well as of regional and long-range transported anthropogenic pollutants were also identified.

- Global circulation modelling indicates that the campaign-based and thus revised VSLS emission scenario now fits much better to the measured tropospheric profile of $CHBr_3$, and CH_2Br_2. It can thus be expected that our understanding of how halogenated VSLS are delivered to the stratosphere will also be further improved in the future.
- Inorganic iodine chemistry, which may efficiently contribute to the oxidation potential of the atmosphere, has only been identified occasionally, i.e. both by ground-based and aircraft-borne observations in the MBL near Semporna, as well as in the free troposphere from the aircraft around the north-eastern tip of Borneo. Signs for an efficient inorganic iodine photochemistry were found as well during the RV Sonne cruise, but the results obtained from three different instruments are still inconclusive to draw a firm picture. However, no signs for an efficient bromine and chlorine photochemistry were found in the MBL or free troposphere, neither during the RV Sonne cruise nor during the ground-based, aircraft-borne or satellite-based observation.
- A persistent though expected feature was the large concentrations of CH_2O and CO produced n the MBL and free troposphere over Borneo, but the concentrations of both pollutants were much lower in the atmosphere over the oceanic margin surrounding Borneo. This observation clearly indicates a rapid photo-oxidation of volatile organic compounds (VOCs) (e.g. isoprene) emitted by the natural and cultivated terrestrial biota.
- During the RV Sonne and local ship cruises, oceanic water was also probed for Zn and Cu concentrations. During a local cruise in the delta of the Sarawak river on Nov. 16, 2011, high Cu (5-55 ppm) but undetectable or low Zn concentrations were found, and conversely high Zn (10-35 ppm) but undetectable or low Cu concentrations were found in the oceanic margins near Kuching on Nov. 19, 2011. The concentrations of both metals in water however, tended to increase towards the open ocean, indicating sources apart from local anthropogenic pollution. Oceanic waters near Kota Kinabalu had undetectable or low Cu but quite high Zn concentrations (15-28 ppm). Zn concentrations were highest at station 4 of the cruise, and Cu at station 5 (2.5 ppm). In Semporna, no Cu (only Station 1 had 0.9 ppm) but again quite high Zn concentrations (12-32 ppm) were found. The highest Zn concentrations were measured near seaweed farms, indicating potential sources there. 210 bacterial isolates were collected during the local boat cruises performed from Kuching, Kota Kinabalu and Semporna (around 70 isolates per cruise). The analyses and interpretation of the filtered samples are ongoing.

Apart from the studies undertaken during the SHIVA SCS campaign of which most data are still being processed and interpreted, to date (Dec. 13) 31 publications related to the SHIVA project appeared. These address the following issues:

A novel method to monitor marine coccolithophore (e.g. haptophytes) blooms from space has been developed [16]. This is relevant since emissions of halogenated VSLS from marine environments are mostly due to the activity of micro- and macro algae, as indicated by previous studies and those performed within SHIVA. Thus, improved space-borne detection of marine micro-biota can help to identify potential sources of VSLS.

Further Brinckmann et al. [4] studied the contribution of halogenated VSLS to Br_y from halogenated VSLS source gas measurements in a source region (Western Pacific) and the tropical tropopause. It was concluded that, at the level of net zero radiative heating (one definition of the mid-TTL), the two brominated VSLS species CH_2Br_2 (average abundance of 1.45 ppt) and

$CHBr_3$ (average abundance of 0.56 ppt) accounted for 90% of the VSLS budget.

Rozanov et al. [13] addressed consolidated retrievals of stratospheric BrO profiles from satellite limb spectroscopy, and the studies of Salawitch et al.[14], and Theys et al. [17] developed new methods on how to attribute measured total atmospheric BrO columns from satellite into stratospheric and tropospheric fractions.

Brioude et al. [5] assessed the ozone depletion potential (ODP) of VSLS emitted in different regions of the world. It was found that ODPs associated with VSLS emissions from the Indian subcontinent are an order of magnitude larger than those from Europe, mid-latitude North America, or East Asia.

To date 10 theoretical studies addressed the transport and transformation of halogenated VSLS in the troposphere. The studies of Hossaini et al. [6], Aschmann et al.[1], and Schofield et al. [15] highlighted that the relevance of marine VSLS emissions for total stratospheric bromine (Br_y) mostly comes through the efficiency of the vertical (convective) atmospheric transport rather than by the amount of VSLS at the surface. Meso-scale transport modelling combined with a detailed scheme for the photochemical degradation of VSLS has not only emphasized the relevance of meso-scale convection, but also some other photochemical and cloud processes enabling an efficient delivery of VSLS and their inorganic product gases to the stratospheric entry level [11, 12]. Hossaini et al. [7] stressed the importance of collocated VSLS sources and convective transport, and Aschmann et al. [2], Krüger et al., [10], Wright et al. [18] and Ashfold et al. [3] emphasized transient times of air mass transport across the tropopause to be key in the cross tropospheric to stratospheric transport. Moreover, the latter studies clearly indicated that most of the air transported from the troposphere into the stratosphere may have its origin in the Western Pacific during the wet season.

A tighter constraint on the magnitude of the photochemical correction when total stratospheric Br_y is calculated from measured stratospheric BrO (i.e. the inorganic method) has been obtained from a comparison of observed and modelled diurnal variations of stratospheric BrO [9]. It is found that stratospheric Br_y is potentially overestimated by up to 1.4 ppt during balloon soundings at high stratospheric NO_x loadings. As a consequence, the present best estimation for the contribution of VSLS bromine to stratospheric Br_y is 4 - 5 ppt (± 2.5 ppt) using the inorganic method, which largely closed the gap to estimations based on the organic method [19].

Finally, in a recent study, Hossaini et al. [8] investigated the role halogenated VSLS may play in a future climate. They found out that in a future climate the delivery of bromine to the stratosphere due to VSLS may increase by 15 – 60%, only considering the more efficient vertical transport in a future climate. However, future VSLS emissions and their contribution to the stratospheric halogen loading may also increase due to (1) increased sea surface temperature and surface winds, and (2) decreased sea level pressure and salinity, e.g. as indicated by simulations of the AOGCM MPI-ESM (for IPCC scenario S-RCP-8.5) and CMAM (by IPCC scenario REF-B2) (Wittke et al., private communication, 2012).

SUMMARY AND OUTLOOK

The present report briefly summarizes the activities and observations made during the SHIVA Western Pacific campaign in fall 2011. Since the data retrieval and interpretation is still ongoing (Dec. 2012), only preliminary findings emerging from the data are reported. These will complement SHIVA related studies that have already been published. Once the ongoing data retrieval and interpretation is more advanced, it can be expected that much more exciting results will emerge from the observations and studies made during the SHIVA SCS campaign. The list of publications related the SHIVA project as well as and digital copies of the publications can be found by visiting the SHIVA project home page http://shiva.iup.uni-heidelberg.de/publications.html.

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