Stratospheric ozone: Halogen Impacts in a Varying Atmosphere
(SHIVA: Grant # 226224)

K. Pfeilsticker¹, M. Dorf¹, B. Sturges², D. Oram², A. Engel³, M. Rex⁴, A. Bracher⁴, M. Roozendael⁵, F. Hendrick⁵, N. Theys⁵, S. Fueglistaler⁶, J. Pyle⁶, B. Quack⁷, K. Krüger⁷, D. Wallace⁷, V. Marecal⁸, V. Catoire⁸, M. Pirre⁸, M. Chipperfield⁹, D. Heard⁹, A. Stohl¹⁰, S. Eckhardt¹⁰, B-M. Sinnhuber¹¹, H. Schlager¹²

(1) Ruprecht-Karls-Universität Heidelberg, Institut für Umweltphysik (UHEI); Germany
(2) University of East Anglia (UEA), United Kingdom
(3) Universität Frankfurt, Institut für Atmosphäre und Umwelt (UFRA), Germany
(4) Alfred-Wegner-Institute for Polar and Marine Research Potsdam and Bremerhaven (AWI), Germany
(5) Belgian Institute for Space Aeronomy (BIRA-IASB), Belgium
(6) University of Cambridge (UCAM), United Kingdom
(7) Leibniz-Institut für Meereswissenschaften an der Universität Kiel (IFM-GEOMAR), Germany
(8) Laboratoire de Physique et Chimie de l'Environnement, Orléans (CNRS)
(9) University of Leeds (UNIVLEEDS), United Kingdom
(10) The Norwegian Institute for Air Research (NILU), Norway
(11) Universität Bremen, Institut für Umweltphysik (UNIHB), Germany
(12) Deutsche Gesellschaft für Luft und Raumfahrt (DLR), Germany
Trend in total stratospheric bromine

Total bromine inferred from balloon-borne observations

Hendrick et al., GRL, 2008

Total bromine over Harestua (60°N) and Lauder (45°S)

Dorf et al., GRL, 2006 with recent updates


The role of VSLS in stratospheric ozone and climate

Chemical and Dynamical Processes Affecting VSLS

Kiruna Sweden
ASA France
Timon Brazil

large-scale transport
stratospheric overworld
28 km
20 km
11 km
cross-tropopause exchange
quasi-horizontal transport
5 km
mid-lat convection
1-2 km
troposphere
PBL/MBL

ozone loss
PG
SG
XO
PG
SG
PG
SG
PG
SG
PG
SG
PG
SG
PG
SG
PG
SG
PG

[Fig. 2.1.3, WMO-2007]

WMO-2007 notation:
VSLS: very short lived species (life times < 6 months)
SG: source gases (e.g. CFC, HCFC, halons, CH3Br, CH3I, CHBr3, CBr2Cl2, etc.)
PG: products gases (e.g. HCl, IO, OIO, BrO, HBr)

Key
VSLS - Very Short-Lived Substances
SG - VSLS Source Gases
PG - Products gases: intermediate organics and inorganic halogen reservoirs
XO - halogen radicals, X=Cl, Br, I
SGI - Source Gas Injection
PGI - Product Gas Injection
TTT - Thermal Tropical Tropopause
TTL - Tropical Tropopause Layer
STT - Secondary Tropical Tropopause
ExTL - Extra-tropical Tropopause Layer
LMS - Lowermost Stratosphere
PBL/MBL - Planetary/Marine Boundary Layer
SHIVA: Some Questions

Q1: What are the relevant processes causing this gap between measured and modeled stratospheric bromine (3 – 8 ppt, mostly likely ~5 ppt)?

Q2: Will this gap - due to influx of brominated VSLS and PGs - change with time?

Q3: What are the present and future impacts of VSLS on the TTL and global ozone?

Q4: What is the case for iodinated species?

Q5: …

Detailed studies of the potential VSLS sources, their strengths, the atmospheric transport and transformation, and the impact on ozone are required!

© WMO-2007 notation:
SG: source gases (CFC, HCFC, halons, CH3Br, CH3I, CHBr3, CBr2Cl2, …
PG: products gases (HCl, IO, BrO, ….
VSLS: very short lived species (life times < 6 months)
SHIVA: Specific scientific objectives

1. The **oceanic emission strengths** of a suite of halogenated source gases

2. The **atmospheric transport and transformation** of the halogenated SGs and PGs

3. The **past, present and likely future trend** of the total halogen burden in the stratosphere

4. The **impact** of long and short-lived **halogenated SGs** and their **inorganic PGs for past, present and future ozone** within the upper troposphere, TTL and global stratosphere
The most relevant question for the HALO deployment within SHIVA!

Where and when act the combination

1. of mostly marine (and less biomass burning) sources of VSLS

and

2. the vertical transport in the atmosphere

efficiently together to deliver VSLS and PGs into and through the TTL eventually into the stratosphere?
Where is the region of interest?

Transport through the TTL – i.e., the layer between 340 K to 400 K is most efficient the largest over the Western Pacific early in the year!

Contour lines are densities of intersections of backward trajectories to points distributed evenly over the 400K surface (~18 km) in the tropics with the 340K surface (~14 km) (i.e. the lower boundary of the TTL), relative to the density of such intersections if spread uniformly over the globe (adapted from Fueglistaler et al., 2004).
When is marine VSLS production the largest?

Spatial distribution of Chlorophyll-a productivity as a proxy for the marine productivity and VSLS emissions!

Annual variation of Chlorophyll-a productivity and possible of marine VSLS emissions!

→ Emissions of VSLS are largest early in the year!
Measurements of bromoform ($\text{CHBr}_3$) at coastal Kunak/Borneo

$\text{CHBr}_3$: GAW tower 12 m (OP3 phase 2)
$\text{CHBr}_3$: mobile GC (OP3 phase 2)

coastal measurements at Kunak

preliminary

On shore wind

Date/time (ticks = 00:00hr local time)

courtesy N. Harris
How are VSLS spatially distributed? -> FLEXPART simulations

**Vertical distribution**

**Horizontal distribution**

Total column of species CHBr3.5 for age class all
Simulation start 20081120. 0 Actual time 20090601.180000
Mean value 0.384E-02
Maximum value 0.527E-01
Minimum value 0.393E-05

Total column of species CHBr3.1 for age class all
Simulation start 20081120. 0 Actual time 20090601.180000
Mean value 0.583E-02
Maximum value 0.337E-01
Minimum value 0.337E-04
When and where is vertical transport into and through the TTL most efficient?

The plots show the relative contribution of air mass fluxes from the ground of 8 equal area boxes (all equal to 20°N – 20°S and in 45° longitude slices) defined in the bottom plot for air reaching at the

- LRM (Lapse Rate Minimum),
- LZRH (Level of Zero Radiative Heating) and
- CPT (Cold Point Tropopause).

In the experiment, a tracer of 2 d life time with 1 ppb in the first 500m was released in the tropical (20°N – 20°S) band. The monthly mean MMR is labeled on the right-side of each bar (Scott Hosking/UCAM priv. comm).

→ Again this study shows that vertical transport into/and through the TTL is most efficient over the Western Pacific early in the year!
A possible to answer the most relevant question (aka where and when are the combination of (marine) sources of VSLS and the vertical transport in the atmosphere efficient to deliver VSLS and product gases into and through the TTL into the stratosphere?)

**VSLS sources are large and atmospheric transport is most efficient**

(1) over the **Western Pacific** during SE monsoon, i.e. in Jan. through March

(2) Other potential important regions and seasons may include those having

  • upwelling oceanic waters, i.e. off-coast tropical Africa (Cap Verde in July to October) or
  • inflow of large and fertile rivers, i.e. Amazon (Dec. through May)

during the wet seasons and/or

(3) biomass burning (e.g., pyroclouds) during the dry season, i.e. Amazon basin, South Eastern Asia, Central Africa in July through November!
Overview over the SHIVA field activities in the Western Pacific in 2011. The colored plot in the background indicate modeled column concentration of CHBr$_3$ in mg/m$^2$ according to GLOBCOLOUR chlorophyll concentration observations and FLEXPART simulations with a tracer of 5d life time. The black dots (1 to 6) mark ground-based observations by the Cambridge $\mu$-Dirac instrument as further detailed in section 4.1.3. The red circles indicate the maximum range of the HALO aircraft for 8h (radius 1200 km) and 1h (radius 3000 km) loitering time in the targeted region for a deployment of the HALO aircraft at Kuching/Borneo (1$^{\circ}$ 30’N, 110$^{\circ}$ 20’E). The yellow ellipse illustrates the area of possible ship cruises.
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<td>Valery Catoire</td>
<td></td>
<td>QCL</td>
<td>N₂O, CH₄, CO, OCS and possibly with 2ⁿᵈ ch either O₃, SO₂, NO₂, or HNO₃</td>
<td>SHIVA</td>
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<td>Andreas Engel</td>
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<td>Christiane</td>
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<td>Manfred Wendisch</td>
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In red: SPP -1294 relevant
Arrangements, Summary & Conclusion

1. The SHIVA project
   • started on July 1, 2009 and will end on June 30, 2012
   • Kick-off meeting was at Heidelberg on July 6/7th, 2009.

2. Measurements of VSLS and other gases will be performed
   • At Cap Verde Islands via ground-based and ship-borne whole air sampling
   • In the Western Pacific via coordinated ground-based, and ship-borne observations in late 2009
   • Combined ship- and aircraft-borne observations in the Western Pacific need to be conducted during the convective season in early 2011, or alternatively 2012.

3. Assessment studies will address
   • global emission inventories & present and future scenarios
   • trends and projections of stratospheric halogen burdens

4. Model work will include
   • process studies of atmospheric transport and pathways (Flexpart, state-of-the-art Eulerian and Lagrangian models of transport and chemistry, Meso-scale modelling e.g. C-CATT-BRAMS)
   • global modelling of the past, present and future (Lagrangian/Eulerian model, CTM, CCM, and calculations)
Invitation

Those who are interested in SHIVA are invited to ....

........ collaborate, contribute, join, et cetera... (upon signing the data protocol)!

→ Thank you!
SHIVA: Working packages

WP 1: Project management (WP leader: Marcel Dorf, IUP Heidelberg)

WP2: Measurements: ground-based, ship-, and aircraft-borne (WP leader: David Oram, UEA)

WP3: Emission inventories and future oceans (WP leader: Birgit Quack, IFM-GEOMAR, Kiel)

WP4: Process studies (WP leader: Markus Rex, AWI)

WP5: Stratospheric halogens analysis and measured trends and projections (WP leader: Björn-Martin Sinnhuber, IUP Bremen)

WP6: Global modeling: past, present and future (WP leader: Stefan Fueglistaler, UCAMB)
Differential Optical Absorption Spectroscopy (DOAS)
Observations from HALO (DFG PF-384 7/1)

K. Pfeilsticker, U. Platt, and T. Wagner(*)
IUP-Heidelberg
University of Heidelberg
(*) now with MPI for Chemistry, Mainz
Time-dependent balloon-borne $O_3$, $NO_2$, and BrO mini-DOAS observations
Time-dependent $O_3$ and $NO_2$ mini-DOAS observations

on LPMA/IASI at Teresina/Brazil on June 30, 2005.

(courtesy: Kritten et al., IUP-Heidelberg)
Mini-DOAS instrument: Technical features

1. Total weight: 47 kg, excl. water 40 kg
2. Dimensions: (mini rack) 43x43x49 cm, (aperture pl.) 10x7\" = 25x18 cm
3. Power: 100 W, 28 VDC
4. Parts:
   - 6 optical spectrometers (2x UV/vis/near-IR in nadir direction and limb scanning) immersed in a pressurized spectrom. housing, ice/water tank for T-control
   - Electronics (PC, T-controller, motor controllers, spectrometer readout electronics)
   - Aperture plate: (1) limb obs. webcam, (2) motors for limb scanning and (3) 6 telescopes
5. Measurement principle: scattered sunlight DOAS combined with RT modelling
6. Detectable species: (1) BrO, ClO OCIO, IO, O₃, HCHO, HONO, SO₂ in the UV;
   (2) I₂, IO, OIO, OBrO, NO₂, O₃, O₄, C₂H₂O₂ in the visible;
   (3) H₂O (g, l, s), O₄, (CO₂, CH₄) in the near-IR

Components of the mini-DOAS instrument

1YMID – spectrometer unit
2-4YMID – electronics unit
5-15YMID – aperture plate
16YMID – mini rack
Mini-DOAS instrument (Oct. 2009)

Aperture Plate
inside

nadir

nadir
telescopes

limb

webcam

telescopes:

limb backward
view

cover for
motor cables

flight
direction

motor 1

motor 2

motor 3

Aperture Plate
Camera + limb Telescopes underneath HALO

HALO miniDOAS Frame

webcam

spectrom. unit

with insulation

and water tank

electronics box 2

Institut für Umweltphysik
University of Heidelberg
1. All mechanical parts are manufactured and close to be assembled
2. Electronics is tested and just being finally assembled
3. Mechanical fitting test of the instrument to the aircraft (Pos 21) is okayed!
4. Major delays is/was with the delivery of the 6 optical spectrometers (from OMT/Ulm), but they should arrive during KW 43 (now !)
5. Software needs to be adapted and finalized
6. Certification documents with OPTIMARE
   • drawings of electrical part and wiring are ready
   • 3D mechanical drawings are a work in progress
   • Certification documents are just being prepared
7. Tbd
   • Tesile/mechanical test
   • EMC test
   • Test and characterization of the instrument in the laboratory

Verlängerung des Antrages zur Teilnahme an den Missionen OMO, TACTS, POLSTRACC, CIRRUS, ACRIDICON, HALO-EO, NARVAL, .. ist vorgesehen!
First Mini-DOAS instrument spectra