Transport Variability of Very Short Lived Substances From the West Indian Ocean to the Stratosphere

Alina Fiehn1,2,3, Birgit Quack2, Christa A. Marandino2, and Kirstin Krüger1

1Meteorology and Oceanography Section, Department of Geosciences, University of Oslo, Oslo, Norway, 2GEOMAR Helmholtz Centre for Ocean Research Kiel, Kiel, Germany, 3Now at Deutsches Zentrum für Luft- und Raumfahrt, Oberpfaffenhofen, Germany

Abstract Halogen- and sulfur-containing compounds are supersaturated in the surface ocean, which results in their emission to the atmosphere. These compounds can be transported to the stratosphere, where they impact ozone, the background aerosol layer, and climate. In this study we calculate the seasonal and interannual variability of transport from the West Indian Ocean (WIO) surface to the stratosphere for 2000–2016 with the Lagrangian transport model FLEXPART using ERA-Interim meteorological fields. We investigate the transport relevant for very short lived substances (VSLS) with tropospheric lifetimes corresponding to dimethylsulfide (1 day), methyl iodide (CH3I, 3.5 days), bromoform (CHBr3, 17 days), and dibromomethane (CH2Br2, 150 days). The stratospheric source gas injection of VSLS tracers from the WIO shows a distinct annual cycle associated with the Asian monsoon. Over the 16-year time series, a slight increase in source gas injection 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. During boreal spring of El Niño, enhanced stratospheric injection 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, strong injection is related to enhanced atmospheric upward motion over the East Indian Ocean and a prolonged Indian summer monsoon season. Related physical mechanisms and uncertainties are discussed in this study.

1. Introduction

Halogens and sulfur in the stratosphere impact the ozone and Junge layers and can, thus, affect the radiation budget and climate (Dvortsov et al., 1999; Myhre et al., 2004; Solomon et al., 1994, 2011). Stratospheric halogen sources include halogenated very short-lived substances (VSLS) emitted from the oceans (Law et al., 2006). Model calculations suggest that marine dimethylsulfide (DMS) emissions contribute to the persistent stratospheric background aerosol layer or Junge layer (Marandino et al., 2013), which is mainly supplied by longer-lived natural sulfur compounds (Crutzen, 1976) and anthropogenic sulfur compounds (Myhre et al., 2004). Natural halogen and sulfur-containing organic compounds originate from chemical and biological sources, such as phytoplankton and macroalgae in the oceans (Carpenter et al., 1999; Moore & Zafiriou, 1994; Quack & Wallace, 2003; Stefels, 2000). Gases, which are emitted to the atmosphere, are defined as VSLS if they have atmospheric lifetimes of less than half a year (Law et al., 2006). In this study, we consider sulfuric and halogenated VSLS, namely DMS, methyl iodide (CH3I), bromoform (CHBr3), and dibromomethane (CH2Br2). These four are the most important oceanic VSLS carriers for halogens and sulfur to the atmosphere, and they represent a large spectrum in the VSLS lifetimes.

Estimates of VSLS emissions from the global oceans are subject to large uncertainties (Carpenter et al., 2014; Lana et al., 2011). Global emission climatologies have been derived from observations and chemistry climate models (bottom-up approach; Butler et al., 2007; Lana et al., 2011; Lennartz et al., 2015; Palmer & Reason, 2009; Quack & Wallace, 2003; Ziska et al., 2013), atmospheric abundances and chemistry climate models (the top-down approach; Liang et al., 2010; Ordóñez et al., 2012; Warwick et al., 2006), and biogeochemical ocean models (Hense & Quack, 2009; Kloster et al., 2006; Stemmler et al., 2015). The delivery of these VSLS to the stratosphere has been the topic of several modeling studies (Hossaini et al., 2010; Kerkweg et al., 2008; Liang et al., 2010; Lennartz et al., 2015; Nielsen & Douglas, 2001; Sheng et al., 2015; Warwick et al., 2006). While the main VSLS source gas injection to the stratosphere occurs over the West Pacific Ocean (Aschmann et al., 2009; Marandino et al., 2013; Tegtmeier et al., 2013), the Asian summer monsoon also is a significant transport pathway (Hossaini et al., 2016; Liang et al., 2014). The few available measurements from the
Indian Ocean showed strong VSLS emissions (Fiehn et al., 2017; Mihalopoulos et al., 1992; Smythe-Wright et al., 2005). Fiehn et al. (2017) emphasized that halogenated VSLS emissions can reach the stratosphere during the summer monsoon season. In addition, the Indian Ocean is a region strongly affected by climate change: the West Indian Ocean (WIO) has been warming faster than any other tropical ocean over the last century (M. K. Roxy et al., 2014) causing a reduction in marine primary production (Roxy et al., 2016), which influences seawater concentrations of DMS and halocarbons (Hepach et al., 2014; Miles et al., 2012).

In the atmosphere, current estimates of tropical tropospheric lifetimes are 1 day for DMS (Barnes et al., 2006; Osthoff et al., 2009), 3.5 days for CH$_3$I, 17 days for CHBr$_3$, and 150 days for CH$_2$Br$_2$ (Carpenter et al., 2014). The delivery of these compounds from the ocean surface to the stratosphere depends on emission strength and vertical transport, because the chemical decay might be faster than the transport time scale. Stratospheric source gas injection of VSLS is connected with fast and high reaching convection above the tropical oceans. The so-called “stratospheric fountain,” the region where tropospheric air enters the stratosphere, is located over the tropical West Pacific from November to March and over the Bay of Bengal and India during the summer monsoon (Newell & Gould-Stewart, 1981). The stratospheric injection is most pronounced over the tropical Pacific and Maritime continent during boreal winter (Fueglistaler et al., 2005; Krüger et al., 2008, 2009), but the Indian summer monsoon is also efficient at transporting boundary layer air masses into the stratosphere (Park et al., 2009; Randel et al., 2010). Several model studies of transport pathways toward the main stratospheric entrance region of the summer monsoon have been carried out. Mostly anthropogenic boundary layer sources were accounted for with chemical transport models (Bergman et al., 2013; Vogel et al., 2015; Vogel et al., 2016) and chemistry climate models (Orbe et al., 2015; Pan et al., 2016). Tzella and Legras (2011) investigated the cross-tropopause transport from convective cloud tops highlighting the importance of localized convective updrafts. Chen et al. (2012) calculated the Asian summer monsoon air mass transport pathways for 2001–2009 with the Lagrangian model FLEXible PARTICle (FLEXPART) and found the main stratospheric entrance regions above the tropical central Indian Ocean, the Bay of Bengal, the South China Sea, and West Pacific. With a chemistry climate model, Liang et al. (2014) simulated a VSLS bromoform maximum above the tropical Indian Ocean at 355 K potential temperature (~13 km) using the emission scenario of Liang et al. (2010). Based on observed emissions of halogenated VSLS from the WIO during July and August 2014 and the model FLEXPART, Fiehn et al. (2017) diagnosed stratospheric injection of CH$_3$I mainly above the equatorial Indian Ocean, while CHBr$_3$ and CH$_2$Br$_2$ reached the stratosphere in the southeastern part of the Asian monsoon anticyclone.

Changes in the transport above the Indian Ocean may originate from changes in the monsoon circulation and its convection. Fast vertical transport in the marine atmosphere, mostly realized through atmospheric deep convection, depends on the convective available potential energy and, thus, heat flux from the ocean connected to the sea surface temperature (SST). Convective activity is often directly related to rainfall and has been used as a precipitation proxy since satellite data became available (Arkin & Ardanuy, 1989). The Indian monsoon rainfall shows changes and variability in the monsoon circulation and convection strength. The leading mode of variability in Indian summer monsoon rainfall is connected with the influence of El Niño–Southern Oscillation (ENSO) on the Walker Circulation (Ding, 2007; Walker, 1924; Wang et al., 2001, 2015; Webster & Yang, 1992). The Asian monsoon system is also experiencing a long-term change (1901–2012) due to greenhouse gas-induced global warming, with a decrease in the land-sea thermal gradient and less summer monsoon rainfall over the central-east and northern regions of India (Roxy et al., 2015), which may lead to reduced convection and VSLS transport to the stratosphere through the summer monsoon.

The contribution of oceanic VSLS to the stratospheric composition and their transport via the Asian monsoon circulation has hardly been studied. In order to better represent natural factors in global chemistry climate and transport models simulating the monsoon system, it is important to also investigate the role of oceanic trace gases, such as VSLS, on atmospheric composition and chemistry. While the atmospheric abundances of many long-lived ozone depleting substances are declining due to the regulation through the Montreal protocol (World Meteorological Organization, 2014), natural VSLS emissions (Ziska et al., 2017) as well as their weighted stratospheric ozone depletion potential (Tegtmeier et al., 2015) could increase in a future climate.

In a previous study, we investigated the stratospheric source gas injection of halogenated VSLS from the WIO during the summer monsoon season. We reported the first measurements of CHBr$_3$ and CH$_2$Br$_2$ from the
Indian Ocean and calculated strong emissions of VSLS from the WIO and their transport to the stratosphere along two pathways, namely the convection above the tropical Indian Ocean and the Indian monsoon circulation (Fiehn et al., 2017). In this follow-up study, we investigate the air mass transport relevant for VSLS source gas injection from the WIO to the stratosphere over the whole year, the seasonal transport cycle, and interannual transport variability. A VSLS tracer with a 1-day lifetime, DMS, was included in this study to investigate transport above the Indian Ocean for the shortest timescale, because the Indian Ocean is an emission hot spot for DMS (Lana et al., 2011). We determine the transport mechanisms and show the relationship between the transport variability above the WIO and the ENSO phase.

2. Methods

2.1. Trajectory Model and Meteorological Data

The air mass transport from the WIO to the stratosphere is calculated with the Lagrangian FLEXPART dispersion model version 9.2 from the Norwegian Institute for Air Research (Stohl et al., 2005). The model includes parameterizations for convection (Forster et al., 2007) and turbulence in the boundary layer and free troposphere (Stohl & Thomson, 1999). FLEXPART uses the model grid of its meteorological input field (Stohl et al., 2005), which is ERA-Interim from the European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis (Dee et al., 2011) in this study. The meteorological input fields were applied at 1° × 1° horizontal and 60 vertical model level resolution, providing 6-hourly air temperature, winds, and specific humidity. A preprocessor, which retrieves the meteorological fields from the ECMWF archives, calculates the vertical wind from spectral data in mass-consistent hybrid coordinates for the FLEXPART runs (Stohl et al., 2005). ERA-Interim input data are retrieved on hybrid coordinates and converted to the internal FLEXPART geometric height. Similar to Fiehn et al. (2017), we start one trajectory per grid point from a 1° × 1° grid at the sea surface within the release area in the tropical WIO (20°S – 10°N, 50°E – 80°E, Figure 1) once every day from 1 January 2000 until 28 February 2016 and calculate them for 3 months forward. The trajectory positions are recorded every 6 hr. First, we determine the path of these trajectories from the ocean surface to an injection height of 17 km, the approximate height of the tropical cold point tropopause (CPT) over the Asian monsoon region (Munchak & Pan, 2014). A sensitivity test on using different thresholds for stratospheric injection is presented in the supporting information. We then calculate the transit time (tt) from the sea surface to the stratosphere for all trajectories that reach 17 km within the simulation time of 3 months. All particles released at the ocean surface carry 100% of the artificial VSLS tracer mass. At 17 km, the remaining mass fraction (f) on each particle is determined by the exponential decay during the transit time (tt) according to the corresponding VSLS lifetimes (τ)

\[ f = e^{-\frac{tt}{\tau}}. \]

We assume tropical tropospheric lifetimes for DMS, methyl iodide, bromoform, and dibromomethane of 1, 3.5, 17, and 150 days, respectively (Barnes et al., 2006; Carpenter et al., 2014; Osthoff et al., 2009). While the number of trajectories reaching 17 km determines the transport of air masses, the fraction of VSLS tracer on the trajectories at 17 km describes the transport on time scales of the VSLS lifetimes. The transport efficiency is defined as the sum of VSLS tracer fractions (fi) entrained on all particles above 17 km divided by the number of particles released (N):

\[ \text{transport efficiency} = \frac{\sum f_i}{N}. \]

The transport efficiency gives a measure of the efficiency of VSLS transport assuming a homogeneous VSLS emission field at the ocean surface. This mass based transport efficiency is comparable to the relative injection used in related studies (Fiehn et al., 2017; Tegtmeier et al., 2012, 2013). The transport efficiency is calculated as monthly mean and is allocated to the month in which the trajectories were initiated to infer the injection strength for oceanic sources at a certain time.

Figure 1. Areas for the definition of climate and circulation indices and the release area for the trajectories and SSTWIO. The indices are the Indian monsoon index (IMI = IMI 1 – IMI 2), dipole mode index (DMI = DMIw – DMIe), and Nino 4 index.
2.2. Climate Indices

In order to investigate the possible influences on seasonal and interannual variability in stratospheric source gas injection of VSLS tracers from the WIO, we use different climate indices (Figure 1) describing the state of the ocean, the atmospheric circulation, and different coupled ocean-atmosphere phenomena. We use ERA-Interim monthly mean SST in our WIO release area (SST\textsubscript{WIO}) to infer transport changes due to local SST anomalies. Above the Indian Ocean, the Indian monsoon circulation dominates the tropospheric circulation. The large scale Indian monsoon circulation is described by the Indian monsoon index (IMI), which is defined as the gradient of zonal wind at 850 hPa between a southern area over the northern WIO (40°E–80°E, 5°N–15°N) and an area over northern India (70°E–90°E, 20°N–30°N; Wang et al., 2001; Wang & Fan, 1999). Daily mean IMI data were provided by Yoshiyuki Kajikawa and Bin Wang on their Monsoon Monitoring Page (Text S1). The strength of the summer monsoon is often described by the amount of rainfall over India. All India rainfall index (AIRI) monthly data are provided by the Indian Institute for Tropical Meteorology for 1871–2014 (Text S1). The SST anomaly between the West and East Indian Ocean, the Indian Ocean dipole (IOD), has been shown to influence convection and rainfall in the Indian Ocean region. The dipole mode index (DMI) describes the status of the IOD and is defined as the difference between SST in the WIO (50°E–70°E, 10°S–10°N) and the east Indian Ocean (90°E–110°E, 10°S–0°, Figure 1; Saji et al., 1999). The DMI is provided by the National Oceanic and Atmospheric Administration (NOAA) State of the Ocean project (Text S1).

We analyze the influence of ENSO on stratospheric injection from the WIO through the monthly SST\textsubscript{Nino 4} averaged over the Nino 4 region (160°E–150°W, 5°N–5°S, Figure 1) from ERA-Interim. We chose the central Pacific Nino 4 over the east Pacific Nino 3.4 region, because central Pacific ENSO events tend to have a greater influence on the Indian Ocean than those in the east Pacific (Kumar et al., 2006).

2.3. Statistical Analysis

We use different statistical analyses to infer trends, variability, and influences between the transport and climate indices in the manuscript. We distinguish the results between intra-annual and interannual variability and long-term changes.

We detrended the 16-year time series of transport efficiency from the WIO to the stratosphere for the calculations of interannual variability and correlations. For this, we first determined the least squares fit to the data and then subtracted the resulting function from them. The significances of the linear trends were computed with a permutation test ($p$ value).

The intra-annual and interannual variability of transport efficiency is investigated with the coefficient of variation, which is defined as the standard deviation of the transport efficiency normalized by its mean. For intra-annual variability, we calculate the intra-annual coefficient of variation from 12 monthly transport efficiency values (Table 1). For interannual variability, we calculate the interannual coefficient of variation from 16 detrended annual values for the annual mean or seasonal means (Table 2). The values of transport efficiency for seasons were calculated from all trajectories started in the designated season. December to February (DJF, boreal winter) is allocated to the year of the starting month, thus trajectories for DJF\textsubscript{2000} were released in December 2000, January 2001, and February 2001. The transport efficiency value of DJF\textsubscript{2015} is the only value that includes the transport efficiency of January and February 2016; all other statistics are based on January 2000 to December 2015.

The correlation of annual cycles and detrended interannual variability of the transport efficiency to the stratosphere with different climate indices are calculated with the correlation coefficient $r$ by Pearson (1895).

<table>
<thead>
<tr>
<th>Table 1</th>
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<tbody>
<tr>
<td>Climatological Annual Mean Transport Efficiency (Percent) and Intra-Annual Coefficient of Variation for Transport From the WIO to the Stratosphere as Displayed in Figure 6a</td>
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<tr>
<td></td>
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<tr>
<td>DMS</td>
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<tr>
<td>---</td>
</tr>
<tr>
<td>Climatological annual mean transport efficiency (%)</td>
</tr>
<tr>
<td>Intra-annual coefficient of variation (unitless)</td>
</tr>
</tbody>
</table>

Note. WIO = West Indian Ocean; DMS = dimethylsulfide.
data were tested for normal distribution. For the correlation of annual cycles, we first calculated the mean annual cycle of transport efficiency for each tracer and for the climate indices from 2000 to 2015 using monthly data. Then a time lag correlation was determined between each tracer’s average annual cycle of transport efficiency and the average annual cycles of the climate indices IMI, AIRI, DMI, SSTWIO, and SSTNino 4. We only show the average correlation over all four VSLS tracers, because the individual correlations are similar. The interannual correlations between the detrended time series of VSLS tracer transport efficiency and climate indices were calculated for annual means as well as for seasonal means of each year. The significance of the correlations was determined with a p value using a Student’s t distribution (Hollander & Wolfe, 1975).

We created maximum injection anomaly composites for all seasons. The 5 years (~30% of the time period) with maximum transport efficiency were determined from the detrended interannual values (Figure S7) and their average was subtracted from the climatological values for the whole 16 years.

### 3. Results

#### 3.1. Case Study 2014

In a previous study we reported about the OASIS (Organic very short-lived Substances and their air sea exchange from the Indian Ocean to the Stratosphere) cruise in the WIO, which included observation-based VSLS emissions and their transport to the stratosphere during Asian summer monsoon (Fiehn et al., 2017). Here we examine the transport during the whole of 2014, the year of the OASIS cruise, to investigate the seasonality of transport from the same release area. The forward trajectories of the first 10 days for every month reflect the different circulation patterns during the seasons (Figure 2). From May to September, the surface summer monsoon circulation is clearly visible above the Indian Ocean. It consists of the southeast trade winds within the release area and the southwest monsoon winds around the northern edge of the release area toward the Indian subcontinent and Bay of Bengal. From October to April, the patterns are less organized. Over the whole year, trajectories released close to the Intertropical Convergence Zone (ITCZ) experience rapid vertical uplift due to convective activity as shown in the supporting information Figure S2 (Schneider et al., 2014). The few northbound and southbound trajectories are rarely lifted to the upper troposphere and lower stratosphere.

We calculated the distribution of VSLS tracers across 1° latitudinal bands to identify the latitude with the highest density of the tracers in 2014. Figure S3 shows this distribution for February and July 2014. Figure 3 shows the latitude of maximum density of VSLS tracers at 8 km, midtroposphere, and 15 km, upper troposphere, for each month in 2014. Overall, the latitude of maximum density of tracers reflects the annual cycle associated with the observed position of the ITCZ and connected precipitation (Schneider et al., 2014). In the midtroposphere, the center of maximum density of tracers moves from 5°S in boreal winter and spring to around 8°N in boreal summer and fall (Figure 3a). In the upper troposphere, the main tracer transport in boreal winter and spring is located as far as 10°S and moves to 20°N in boreal summer, depending on the lifetime of the tracer

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**Table 2**

<table>
<thead>
<tr>
<th></th>
<th>DMS</th>
<th>CH3I</th>
<th>CHBr3</th>
<th>CH2Br2</th>
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<tbody>
<tr>
<td><strong>Climatological transport efficiency (%)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Annual mean</td>
<td>0.34</td>
<td>0.68</td>
<td>2.13</td>
<td>6.50</td>
</tr>
<tr>
<td>Seasonal means</td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>DJF</td>
<td>0.34</td>
<td>0.66</td>
<td>2.07</td>
<td>6.52</td>
</tr>
<tr>
<td>MAM</td>
<td>0.64</td>
<td>1.19</td>
<td>3.04</td>
<td>7.66</td>
</tr>
<tr>
<td>JJA</td>
<td>0.21</td>
<td>0.51</td>
<td>1.96</td>
<td>5.91</td>
</tr>
<tr>
<td>SON</td>
<td>0.17</td>
<td>0.37</td>
<td>1.47</td>
<td>5.70</td>
</tr>
<tr>
<td><strong>Interannual coefficient of variation (unitless)</strong></td>
<td></td>
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<tr>
<td>Annual mean</td>
<td>0.14</td>
<td>0.14</td>
<td>0.12</td>
<td>0.08</td>
</tr>
<tr>
<td>Seasonal means</td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>DJF</td>
<td>0.33</td>
<td>0.31</td>
<td>0.23</td>
<td>0.13</td>
</tr>
<tr>
<td>MAM</td>
<td>0.17</td>
<td>0.15</td>
<td>0.10</td>
<td>0.07</td>
</tr>
<tr>
<td>JJA</td>
<td>0.33</td>
<td>0.21</td>
<td>0.12</td>
<td>0.07</td>
</tr>
<tr>
<td>SON</td>
<td>0.42</td>
<td>0.38</td>
<td>0.27</td>
<td>0.13</td>
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</tbody>
</table>

Note. DJF = December–February; MAM = March–May; JJA = June–August; SON = September–November.
In July, the distance between the main pathways of the two longer-lived tracers (CHBr₃ and CH₂Br₂, 20°N) and the two shorter-lived tracers (DMS and CH₃I, 8°N) in the upper troposphere is about 12° in latitude (see also Figure S3). This splitting of the main transport pathways according to tracer lifetime confirms the two main summer monsoon transport pathways found by Fiehn et al. (2017): (1) the very fast uplift over the Indian Ocean for CH₃I and (2) the slower transport to the north and lifting with the Asian

Figure 2. Ten-day forward trajectories released at the sea surface of the tropical WIO (red box) in 2014. The trajectories are plotted translucently, so that a more intense blue color displays a higher density of trajectories and, thus, a more important transport pathway.

(Figure 3b). In July, the distance between the main pathways of the two longer-lived tracers (CHBr₃ and CH₂Br₂, 20°N) and the two shorter-lived tracers (DMS and CH₃I, 8°N) in the upper troposphere is about 12° in latitude (see also Figure S3). This splitting of the main transport pathways according to tracer lifetime confirms the two main summer monsoon transport pathways found by Fiehn et al. (2017): (1) the very fast uplift over the Indian Ocean for CH₃I and (2) the slower transport to the north and lifting with the Asian

Figure 3. Annual cycle of latitude of maximum density of WIO to stratosphere transport at (a) 8 km and (b) 15 km altitude in 2014.
monsoon circulation for CHBr₃ and CH₂Br₂. The annual cycle (Figure 3b) also shows that the two pathways only exist during boreal summer. During the onset of the summer monsoon, in April and May, the CHBr₃ and CH₂Br₂ tracer pathways shift northward about a month earlier than for the DMS and CH₃I tracers. After the end of the summer monsoon in November the pathways of the two longer-lived tracers shift southward a month earlier. This difference in the seasonal shift is not caused by the monsoon circulation but is an effect of the different lifetimes and the plotting of the transport characteristics over the whole lifetime at the release time of the trajectories (section 2.1). The VSLS tracers released in April are influenced by the premonsoonal circulation in April if they have short lifetimes (DMS and CH₃I), and also by the onset of the monsoon in May if the lifetimes are longer (CHBr₃ and CH₂Br₂).

With the change in tropospheric circulation in the tropics over the year, the stratospheric injection region also varies. Figure 4 shows where the CHBr₃ tracer from the WIO first reaches 17 km for every month of 2014. Injection regions for DMS, CH₃I, and CH₂Br₂ tracers can be found in the supporting information (Figures S4–S6). From May to September 2014, during the summer monsoon, the stratospheric injection region lies
above the Bay of Bengal, northern India, and East Asia for CHBr$_3$ and CH$_2$Br$_2$. The DMS and CH$_3$I tracer summer monsoon injection at 17 km is further south at the northern edge of the release area, around 10$^\circ$N. From October to April 2014, during the winter monsoon, the main stratospheric injection region of all tracers lies above the release box in the WIO. This is in accordance with their maximum transport density in the upper troposphere found in Figure 3b. During boreal summer (June–August, JJA) and fall (September–November, SON) 2014, the transport pathway from the WIO release area to the main uplift and injection region is much longer than during the rest of the year (Figure S2). This leads to more decay of the VSLS tracers and a minimum in transport efficiency in boreal summer and fall (Figures 4 and S4–S6). The maximum transport efficiency from the WIO to the stratosphere for all tracers occurs during boreal spring (March–May, MAM) 2014.

3.2. Climatology

The monthly transport efficiencies from 2000 to 2016 of the VSLS tracers from the WIO to the stratosphere show a distinct annual cycle, interannual variability, and long-term changes (Figure 5). We analyze these variations in this section.

3.2.1. Annual Cycle

The climatological annual cycles of transport efficiency for all four VSLS tracers display maximum injection in boreal spring and minimum injection in boreal fall (Figure 6a). The climatological annual mean transport efficiency and their intra-annual variation are summed up in Table 1. As expected, the climatological annual mean transport efficiency is highest for the longest-lived tracer, CH$_2$Br$_2$, and lowest for the shortest-lived tracer, DMS. The intra-annual coefficient of variation shows that the DMS annual cycle has the highest relative amplitude and the CH$_2$Br$_2$ cycle shows the smallest amplitude. There is, thus, an inverse relationship between the lifetime of tracers and the amplitude of their annual cycles of transport efficiency.

We conducted a time lag correlation between the climatological annual cycles of VSLS tracer transport efficiency (Figure 6a) and the climatological annual cycles of IMI, AIRI, DMI, SSTWIO, and SSTNino 4 climate indices (Figure 6b). The average time lag correlation coefficients for all VSLS tracers are displayed in Figure 6c. Without time lag, the correlation coefficient of transport efficiency with IMI, AIRI, DMI, and SSTNino 4 is between $-0.3$ and $-0.5$ with a significance threshold of $0.58$. The SSTWIO shows a significant positive correlation of 0.8 without lag. For AIRI, IMI, and SSTNino 4, we find significant maximum negative correlations with a time lag of 2 months, maximum positive correlations with a lag of 7 to 9 months. This means that the maximum of injection occurs 2 months after the minimum in monsoon circulation and rainfall over India. Significant correlations with DMI have a minimum without time lag and a maximum at 5 months, thus maximum stratospheric injection occurs at the time of year with the weakest DMI. The annual cycle of the SSTWIO has highest significant correlations to the stratospheric injection without time lag and smallest correlations at 8 months. Thus, the maximum stratospheric injection coincides with the time of highest SST in the WIO.

3.2.2. Interannual Variability

The Asian monsoon circulation, as well as the convection strength over the Indian Ocean, is subject to interannual variability, which might be reflected in the VSLS transport efficiency from the WIO to the stratosphere.
To determine interannual variability of transport efficiency, we calculated the coefficient of variation from the detrended time series shown in the supporting information in Figure S7. The interannual coefficient of variation for the annual mean is less than for the seasonal means (Table 2). The seasons with high interannual variability are boreal fall and winter, while variability is lowest during maximum injection in boreal spring.

There are differences between the VSLS tracers as well. The shortest-lived tracer (DMS) is most variable on interannual scales, and interannual variability decreases as lifetime increases. When comparing the interannual variability (Table 2) to the annual cycle (Table 1), we find that interannual variability is weaker than the amplitude of annual cycle.

In order to investigate oceanic and atmospheric factors that influence interannual VSLS transport variability, we calculated correlations between the detrended 16-year interannual time series of VSLS tracer transport efficiency (Figure S7) and the climate indices (section 2.2) describing the state of the ocean in the release area (SSTWIO), the Indian monsoon strength (IMI), the state of the IOD (DMI), and the central Pacific ENSO conditions (SSTNino 4; Table 3). We discuss only those relationships that show a significant correlation (bold font in Table 3).

The SSTWIO in the WIO release area shows a positive interannual correlation with transport efficiency in MAM for all tracers, in DJF and JJA for DMS and CH$_3$I and for DMS in the annual mean. These positive correlations imply that higher SST in the release area during the mentioned seasons is connected with enhanced transport to the stratosphere. Correlations between IMI and the transport efficiency are low and not significant. The interannual correlations between the DMI and transport efficiency are only significant in JJA for DMS and CH$_3$I. The SSTNino 4 has the strongest relationship with transport efficiency of CH$_2$Br$_2$ and CHBr$_3$ from the WIO to the stratosphere during boreal fall, when negative SST anomalies in the central equatorial
Table 3  
Correlations Between the Detrended Interannual Transport Efficiency From the WIO to the Stratosphere as Annual and Seasonal Means and SST_{WIO}, the IMI, the DMI, and SST_{Nino 4}.

<table>
<thead>
<tr>
<th></th>
<th>Annual mean</th>
<th>DJF</th>
<th>MAM</th>
<th>JJA</th>
<th>SON</th>
</tr>
</thead>
<tbody>
<tr>
<td>SST_{WIO} DMS</td>
<td>0.55</td>
<td>0.66</td>
<td>0.85</td>
<td>0.53</td>
<td>0.39</td>
</tr>
<tr>
<td>CH3I</td>
<td>0.44</td>
<td>0.60</td>
<td>0.85</td>
<td>0.55</td>
<td>0.28</td>
</tr>
<tr>
<td>CHBr3</td>
<td>0.14</td>
<td>0.43</td>
<td>0.71</td>
<td>0.48</td>
<td>0.08</td>
</tr>
<tr>
<td>CH2Br2</td>
<td>−0.02</td>
<td>0.16</td>
<td>0.52</td>
<td>0.38</td>
<td>0.06</td>
</tr>
<tr>
<td>IMI DMS</td>
<td>−0.12</td>
<td>−0.05</td>
<td>−0.44</td>
<td>−0.06</td>
<td>−0.16</td>
</tr>
<tr>
<td>CH3I</td>
<td>0.06</td>
<td>−0.02</td>
<td>−0.42</td>
<td>0.00</td>
<td>−0.01</td>
</tr>
<tr>
<td>CHBr3</td>
<td>0.44</td>
<td>0.02</td>
<td>−0.26</td>
<td>0.17</td>
<td>0.25</td>
</tr>
<tr>
<td>CH2Br2</td>
<td>0.41</td>
<td>−0.01</td>
<td>−0.11</td>
<td>0.23</td>
<td>0.44</td>
</tr>
<tr>
<td>DMI DMS</td>
<td>0.29</td>
<td>0.36</td>
<td>−0.24</td>
<td>0.75</td>
<td>0.48</td>
</tr>
<tr>
<td>CH3I</td>
<td>0.26</td>
<td>0.38</td>
<td>−0.14</td>
<td>0.68</td>
<td>0.28</td>
</tr>
<tr>
<td>CHBr3</td>
<td>0.08</td>
<td>0.34</td>
<td>−0.12</td>
<td>0.41</td>
<td>−0.08</td>
</tr>
<tr>
<td>CH2Br2</td>
<td>0.17</td>
<td>0.10</td>
<td>−0.14</td>
<td>0.18</td>
<td>−0.25</td>
</tr>
<tr>
<td>SST_{Nino 4} DMS</td>
<td>0.47</td>
<td>0.32</td>
<td>0.69</td>
<td>0.08</td>
<td>−0.20</td>
</tr>
<tr>
<td>CH3I</td>
<td>0.21</td>
<td>0.27</td>
<td>0.64</td>
<td>−0.04</td>
<td>−0.39</td>
</tr>
<tr>
<td>CHBr3</td>
<td>−0.18</td>
<td>0.12</td>
<td>0.49</td>
<td>−0.18</td>
<td>−0.67</td>
</tr>
<tr>
<td>CH2Br2</td>
<td>−0.34</td>
<td>−0.08</td>
<td>0.38</td>
<td>−0.21</td>
<td>−0.72</td>
</tr>
</tbody>
</table>

Note: Bold numbers are significant at the 95% level. DJF = December–February; MAM = March–May; JJA = June–August; SON = September–November; WIO = West Indian Ocean; DMS = dimethylsulfide; IMI = Indian monsoon index; DMI = dipole mode index; SST = sea surface temperature; SST_{Nino 4} = SST in the Nino 4 Region; SST_{WIO} = SST in the WIO.

Pacific relate to enhanced stratospheric injection. In MAM, positive correlation coefficients hint at a relationship between positive SST anomalies in the Nino 4 region and enhanced stratospheric injection from the WIO.

Based on these interannual correlations, we calculated SST and vertical velocity anomaly composite maps for the 5 years with maximum transport efficiency after detrending (Figure 8). The maximum transport efficiency years for each season are listed in Table 4. The composites show anomalies that lead to more transport from the WIO to the stratosphere during each season. The DJF composite for SST illustrates positive and negative SST anomalies in the Indian Ocean but maximum anomaly in the equatorial Pacific. Vertical velocity anomalies in DJF include slower upward movement over the Indian Ocean but faster upward movement over the central equatorial Pacific. The MAM composites reveal enhanced SST and upward air motion over the Indian Ocean, resulting in the significant MAM-SST_{WIO} correlation for all tracers in Table 3. In the Pacific Ocean, the composite indicates positive SST anomalies along the equator, resembling the positive correlation with SST_{Nino 4} in MAM.

The JJA composites reflect the positive IOD pattern in the Indian Ocean, related to our positive correlation between DMI and transport efficiency for DMS and CH3I. Vertical velocities show enhanced upward movement over the central Indian Ocean. In SON, the SST anomaly structure over the Indian Ocean is divided, with lower anomalies in the WIO and higher anomalies in the East Indian Ocean. The tropical and subtropical Pacific Ocean displays negative SST anomalies over a large region, reflecting the negative correlations of transport efficiency with SST_{Nino 4} in Table 3. Vertical velocities in SON show enhanced uplift over the central tropical Indian Ocean, the Bay of Bengal, Bangladesh, and the South China Sea. Overall, the derived maximum transport efficiency composites reflect the influence of ENSO events in particular for DJF and SON well (see Table 4).

3.2.3. Long-Term Changes

The interannual time series of VSLS tracer transport efficiency shows a slight increase over the 16 years for all tracers and seasons (Figure 7). The decadal increase in transport efficiency is noted in Table 5. The long-term changes of transport efficiency are most significant for the two longer-lived VSLS CHBr3 and CH2Br2. The detrended interannual time series of transport efficiency are shown as anomalies in Figure 57.

4. Discussion

4.1. Annual Cycle

The transport of VSLS tracers from the WIO to the stratosphere experiences a distinct annual cycle modulated by the Indian monsoon winds, movement of convection connected with the ITCC (Lawrence & Lelieveld, 2010; Schneider et al., 2014), and the SST in the tropical WIO and Central Pacific Ocean (Figure 3). During boreal summer, the maximum density of transport at 8-km height (Figure 3a) occurs over southern India and the Bay of Bengal (10°N), farther south than the climatological position of the rainfall maximum. This can probably be explained by the center of our release area being located to the south of the equator. At 15-km height, the difference between the location of transport of DMS and CH3I versus CHBr3 and CH2Br2 (Figure 3b) in boreal summer can be related to their different lifetimes. The two shorter-lived tracers need very fast transport close to the release area to reach the stratosphere. The two longer-lived VSLS tracers survive long enough to be transported on a pathway farther to the north, with the Indian summer monsoon convection located over the Indian subcontinent. This transport pathway has been described previously by Fiehn et al. (2017) for 2014.

With the seasonal change of atmospheric circulation above the WIO, the stratospheric source gas injection regions change as well (Figure 4). For DMS and CH3I, the main stratospheric injection region remains
above the release area in the WIO all year, because the lifetimes of these tracers are too short for long range horizontal transport and injection (Figures S4 and S5). The region of main injection of CHBr3 and CH2Br2 tracers to the stratosphere (Figure 4 and S6) moves from the release area in DJF and MAM toward the Bay of Bengal, northern India, and Southeast Asia in JJA and SON (section 3). The convection over the Bay of Bengal, northern India, and Bangladesh has previously been shown to be a major pathway from the Indian and Tibetan Plateau boundary layer and troposphere to the stratosphere during boreal summer (Bergman et al., 2013; Pan et al., 2016; Tissier & Legras, 2016). Transport and stratospheric injection from the WIO with the summer monsoon circulation only play a role for VSLS with lifetimes on the order of CHBr3 (17 days) or longer. Source regions closer to the monsoon convection, especially the Bay of Bengal, would be

Table 4

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>MAM</td>
<td>2001</td>
<td>2003</td>
<td>2005</td>
<td>2010</td>
<td>2012</td>
</tr>
</tbody>
</table>

Note: ENSO events based on the ocean Niño index from NCEP/CPC are highlighted with red for El Niño and blue for La Niña; note that “DJF 2002” stands for D2002-JF2003 ff. DJF = December–February; MAM = March–May; JJA = June–August; SON = September–November; ENSO = El Niño–Southern Oscillation; CPC = Climate Prediction Center; NCEP = National Centers for Environmental Prediction.

Figure 8. Composites of SST and vertical wind (at 200 hPa) anomaly for the detrended five maximum injection years (see Table 4) from ERA-Interim monthly means. Negative values of vertical velocities (in Pa/s) relate to enhanced uplift. DJF = December–February; MAM = March–May; JJA = June–August; SON = September–November; SST = sea surface anomaly.
The interannual variability of transport efficiency from the WIO to the stratosphere shows a strong relationship with tropical SST both in the Indian and in the Pacific Ocean (Table 3). The SST in the tropical WIO is an important factor influencing the transport efficiency, especially its annual cycle. The annual cycle of our transport efficiency can be related to the seasonality of SST in the WIO and the Indian monsoon circulation. This is supported by the significant positive correlation with the SST-WIO annual cycle without time lag and negative correlation with IMI and AIRI annual cycles with a time lag of only 2 months (Figure 6). Chen et al. (2012) diagnose Indian Ocean and West Pacific boundary layer to tropopause transport time scales for convective transport of 1–2 days. This fits to our SST-WIO annual cycle correlation without time lag and underlines that the SST in the WIO has an immediate effect on the local convection and uplift and, thus, also on the transport to the stratosphere. Chen et al. (2012) also find a large scale circulation transport timescale of 1–7 weeks, which fits the IMI/AIRI annual cycle correlation with a 2 month time lag. This time lag is caused by the time delay between the circulation change in the lower troposphere, where the indices are calculated, and the establishment of the transport pathway up to the stratosphere after the shift in the large-scale atmospheric circulation pattern.

Finally, the choice of the release area above the Indian Ocean influences the transport efficiency, especially its annual cycle. The annual cycle is strongly connected to the movement of the ITCZ over the Indian Ocean with the highest transport efficiency when the ITCZ is directly over the VSLS release area. Nevertheless, other sub-release areas within the Indian Ocean would also show a distinct annual cycle modulated by the Asian monsoon.

### 4.2. Interannual Variability

The interannual variability is much smaller than the annual cycle (section 3.2). Furthermore, the interannual variability for seasonal means is higher than for annual means, probably because temporal shifts in the pronounced annual cycle cause large variations from year to year (Table 2). An early onset or decline of the summer monsoon can cause transport shifts between the seasons, but the annual mean transport might remain the same.

The interannual variability of transport efficiency from the WIO to the stratosphere shows a strong relationship with tropical SST both in the Indian and in the Pacific Ocean (Table 3). The SST in the tropical WIO

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Table 5
Increase of VSLS Tracer Transport Efficiency From the West Indian Ocean to the Stratosphere During 2000–2015 (in Percent per Decade) for Annual and Seasonal Means

<table>
<thead>
<tr>
<th>Tracer</th>
<th>Annual mean</th>
<th>DJF</th>
<th>MAM</th>
<th>JJA</th>
<th>SON</th>
</tr>
</thead>
<tbody>
<tr>
<td>DMS</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>CH₃I</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>CHBr₃</td>
<td>0.04</td>
<td>0.07</td>
<td>0.03</td>
<td>0.03</td>
<td>0.05</td>
</tr>
<tr>
<td>CH₂Br₂</td>
<td>0.08</td>
<td>0.10</td>
<td>0.07</td>
<td>0.06</td>
<td>0.08</td>
</tr>
</tbody>
</table>

Note: Bold numbers mark significant changes at 95% level. DJF = December–February; MAM = March–May; JJA = June–August; SON = September–November; DMS = dimethylsulfide; VSLS = very short lived substances.
enhances the transport to the stratosphere for the shortest-lived tracers DMS and CH$_3$I during most seasons. For CHBr$_3$ and CH$_2$Br$_2$ the correlation is strongest in MAM, when the fast uplift is located over the equatorial WIO, but negligible in SON, when the main uplift of these tracers occurs above the Bay of Bengal far away from our WIO release region. The positive relationship between the local SST$_{WIO}$ and transport efficiency reveals that the convective energy available from the warm ocean plays a role when the injection occurs directly above the release area. The positive DMI-transport efficiency correlation (impact of the IOD) for the two shortest-lived tracers in JJA supports this assumption, because a positive IOD also includes high SST anomalies in the WIO. Since DMS and CH$_3$I are not entrained via the Indian summer monsoon transport pathway (see section 3.1 and Fiehn et al., 2017), the local influence of the IOD on uplift plays a role.

In boreal spring, the SST$_{Nino 4}$ in the equatorial central Pacific has a positive relationship with stratospheric injection suggesting that El Niño-like conditions enhance stratospheric injection from the WIO. Figure 9 depicts how El Niño events influence the SST and tropospheric vertical velocities above the Indian and Pacific Oceans through perturbations in the Walker circulation. SST warming in the central-to-east equatorial Pacific (El Niño) locally generates enhanced ascent of air, anomalous subsidence over the tropical East Indian and West Pacific Ocean. It also leads to enhanced uplift over the WIO (Wang et al., 2001; Webster & Yang, 1992), which is the main injection region in boreal spring. Additionally, El Niño leads to a basin-wide warming of the Indian Ocean SST (Ju & Slingo, 1995; Schott et al., 2009) and further enhanced uplift over the WIO (M. K. Roxy et al., 2014; Yu & Rienecker, 1999). In our study, El Niño conditions and a warm WIO surface increase the delivery of oceanic WIO VSLS tracers to the stratosphere during boreal spring.

In boreal fall (SON), La Niña-like SST anomalies in the tropical central and East Pacific cause stronger than normal stratospheric injection over the East Indian Ocean (Table 3 and Figure 8). The main uplift and injection regions of sources from the WIO are above the central and northeast Indian Ocean (Figure 4), and La Niña-like SST anomalies in the east Pacific enhance upward movement in this region (Figure 9). Thus, La Niña events strengthen the monsoon convection and uplift over the Indian continent (Wang et al., 2015). The negative SST$_{Nino 4}$ anomalies in boreal fall may extend the summer monsoon season and its strong convection (Goswami & Xavier, 2005; Xavier et al., 2007), bolstering and prolonging the established pathway by the Asian summer monsoon and enhancing stratospheric VSLS injection. This analysis highlights that the shift of stratospheric injection from the WIO in boreal spring to the central and northeast Indian Ocean in boreal fall causes opposing effects of El Niño and La Niña on transport efficiency during these seasons.

The maximum injection years in Table 4 reflect the connection between ENSO and the injection strength. In DJF and MAM maximum injection occurs during El Niño, while in JJA and SON La Niña events dominate the maximum injection.

4.3. Long-Term Changes

The VSLS transport efficiency from the WIO to the stratosphere shows a slight increase from 2000 to 2015 for all tracers and seasons (Figure 7). This is not supported by Hossaini et al. (2016), who investigated global long-term changes in the stratospheric source gas injection of brominated VSLS from 1993 to 2012 with global chemistry transport and chemistry climate models. The global study by Tegtmeier et al. (2015) reports overall no change for the updraught mass flux between 250 and 80 hPa of ERA-Interim data during 1979–2013 but
an increase from 2000 onward. Chen et al. (2012) calculated 9 years (2001–2009) of boreal summer air mass transport in the Asian monsoon region but could not detect any significant correlations between boundary layer source variability and ENSO. Our time series is too short, however, to diagnose a trend, but detecting an increase in transport efficiency for all seasons suggests a general circulation change in the Indian Ocean and Asian monsoon region during the 16-year time period. Indeed, an increase in precipitation over northeastern India was reported during the time interval of our transport study (Latif et al., 2016), hence an increase of convection in our transport area, which can explain the increase in stratospheric injection. However, the same authors detect an overall decrease in northeastern Indian rainfall during 1950s–2010s, potentially impacting the VSLS transport efficiency. Roxy et al. (2017) report about a threefold increase in widespread extreme rain events over central India during 1950–2015, while the mean precipitation has declined with the weakening of the monsoon circulation. The connection between precipitation and intense vertical transport to the stratosphere may explain our increasing transport efficiency.

4.4. Uncertainties

Our conclusions are subject to uncertainties resulting from the calculation and analysis of the trajectories and correlation methods used. The trajectory calculations depend on the Lagrangian model FLEXPART 9.2 and the reanalysis data ERA-Interim used to analyze the air mass transport. Convection generally takes place on scales smaller than the reanalysis model resolution. Therefore, FLEXPART includes a convection scheme to parameterize representative vertical displacement (Forster et al., 2007). Using also FLEXPART/ERA-Interim trajectory calculations, with observation-based VSLS emissions, Tegtmeier et al. (2013) and Fuhlbrügge et al. (2016) were able to reproduce vertical VSLS distributions measured during several aircraft campaigns in the tropics. In a backward trajectory study to investigate the source regions of the Asian monsoon anticyclone, Bergman et al. (2013) compared results including different reanalysis data sets and resolutions and the use of diabatic and kinematic vertical velocities. The largest influence had the resolution of the fields, hinting that very deep, small-scale convective cells are important for the transport of air from the ocean to the stratosphere.

The number of trajectories reaching the stratosphere depends on the definition of the boundary between troposphere and stratosphere. In the tropics, the CPT is often used to describe this boundary (Carpenter et al., 2014). CPT height lies around 17 km in the tropics; it varies slightly with season and latitude and is lifted to about 17.7 km in the Asian monsoon region during boreal summer (Munchak & Pan, 2014, and references therein). During the OASIS campaign in boreal summer in the subtropical and tropical WIO, an average CPT height of 17 km was determined with 2–8 radiosonde launches daily (Fiehn et al., 2017). ERA-Interim reveals an annual average CPT height between 30°S and 30°N of 16.25 km for the year 2014, which is varying seasonally and regionally (supporting information of Fiehn et al., 2018).

We conducted a sensitivity test for the year 2014 to investigate the effect of 17 km as boundary to the stratosphere compared to the CPT and 18 km as injection thresholds (Table 6; Figure S1 and Text S2). We also added the transport efficiency for air masses, which does not consider any decay but is restricted to the simulation time of 90 days (Table 6). The CPT height was taken from the ERA-Interim 60 level model grid with a vertical resolution of about 1 km in the tropical tropopause region. As Figure S1 from Fiehn et al. (2018) shows the ERA-Interim CPT is lower for the JJA mean of 2014 compared to the OASIS radiosonde measurements (Fiehn et al., 2017) and also for the long-term annual mean compared to Global Positioning System radio occultation observations (not shown here). In total, the sensitivity test reveals that 17 km is a more

| Table 6 Annual Mean Transport Efficiency (in Percent) for VSLS Tracers and Air Masses From the WIO to the Tropical Stratosphere (30°S–30°N) Using Different Stratospheric Boundaries as the CPT, 17 km, 18 km, and 17 km With the FLEXPART Convection Scheme Switched Off (17 km No Convection) |
|-----------------|----------|----------|----------|----------|----------|
|                 | DMS      | Methyl iodide | Bromoform | Dibromomethane | Air masses |
| CPT             | 0.33     | 1.03      | 4.15      | 8.34      | 10.60     |
| 17 km           | 0.24     | 0.72      | 2.89      | 5.96      | 7.64      |
| 18 km           | 0.06     | 0.19      | 1.08      | 2.65      | 3.56      |
| 17 km No convection | 0.00  | 0.01      | 1.04      | 3.83      | 5.52      |
conservative threshold than the CPT, while the seasonality is maintained. The VSLS tracer injection at 18 km is much smaller than at 17 km because the transit time to 18 km is much longer (not shown here).

Table 6 also displays the annual mean transport efficiency for the four VSLS at 17 km with the convection scheme switched off (called "no convection") for the year 2014. The two shorter-lived VSLS DMS and CH$_3$I barely reach the stratosphere without convection. The injection of CHBr$_3$ and CH$_2$Br$_2$ is reduced by 65% and 35%, respectively. As we can also see from Figure S1 the seasonal cycle of transport efficiency changes its characteristics when turning off the convection scheme, revealing the maximum transport efficiency during July and December. Thus, the consideration of convection seems to be the key for the transport efficiency results in our study and in Tissier and Legras (2016).

Trends in tropical tropopause height (Seidel and Randel, 2006) could induce trends in stratospheric delivery of VSLS using a static boundary of 17 km. A higher, respectively lower, tropical tropopause may lead to faster, respectively slower, transport from the surface to 17-km height as it means that the VSLS tracers would still be in the troposphere, respectively have already reached the stratosphere. As our analysis is based on ERA-Interim, which includes reanalysis uncertainties with regard to the tropical tropopause height in itself, that is, due to changes of assimilated observations (Fjiiwara et al., 2017) within the considered time period of 16 years, we evaluate using 17-km altitude as a conservative estimate here.

Our study is based on the homogeneous release of tracers from the ocean surface. In reality oceanic VSLS emissions exhibit both spatial and temporal variations (Lana et al., 2011; Lennartz et al., 2015; Ziska et al., 2013). Therefore, our analysis only detects influences of circulation variability on the transport to the stratosphere but neglects variability in the strength of emissions and their influence on the absolute delivery of VSLS to the stratosphere, which will be addressed in a follow-up study.

The total delivery of oceanic bromine, iodine, or sulfur to the stratosphere is based on the injection of the source gases and their soluble product gases. In this study, we consider oceanic source gases and their transport only. Their stratospheric injections are generally enhanced with enhanced vertical uplift (Hossaini et al., 2010). The total stratospheric VSLS bromine delivery of 2–8 pptv is estimated to consist by half of source gas injection and half of product gas injection (Carpenter et al., 2014).

The deductions about the relationships between interannual variation of transport efficiency and SST in the Indian and Pacific Ocean depend on the considered time period. The 16 years time series is too short to detect significant long-term trends in the WIO transport to the stratosphere. However, this study is the longest transport time series calculated and analyzed for intra-annual and interannual variability of Lagrangian transport from the WIO through the Asian Monsoon circulation to the stratosphere so far. The variability in conclusions on the development of transport to the stratosphere is large (see section 4.3). Since ENSO shows the strongest influence on transport efficiency from the WIO to the stratosphere, it is important to cover a period long enough to represent both ENSO phases well. Our chosen time period 2000–2016 is characterized by a clustering of La Niña events and a negative phase of the Pacific Decadal Oscillation (PDO; Mantua & Hare, 2002). The PDO has a very similar impact on the Indian summer monsoon rainfall to ENSO events, with a negative PDO phase related to an increase in summer monsoon rainfall and vice versa (Krishnan & Sugi, 2003). Thus, the reduced global climate variability due to the prevailing negative PDO phase might make it harder to find significant transport-climate variability relationships. A longer time series including more El Niño events and a positive PDO phase is needed for future studies.

5. Summary and Conclusions

A 16-year time series of transport from the tropical WIO to the stratosphere is analyzed using VSLS tracers with lifetimes corresponding to DMS (1 day), CH$_3$I (3.5 days), CHBr$_3$ (17 days), and CH$_2$Br$_2$ (150 days). The transport efficiency, the fraction of tracer entrained above 17 km, of all four VSLS tracers shows a distinct annual cycle with a maximum of source gas injection in boreal spring and a minimum in boreal fall. This is caused by a seasonal change in the main transport pathway due to the reversal of lower tropospheric winds above the WIO connected to the Asian monsoon circulation. With the onset of the summer monsoon in May, the region of main upward transport of VSLS tracers moves from south of the equator to about 20°N. With the main vertical uplift, the main stratospheric injection region of the longer-lived VSLS tracer CHBr$_3$ and CH$_2$Br$_2$ shifts to the Bay of Bengal and northern India, lengthening the transport pathway from the WIO to the
stratosphere and causing a decrease in transport efficiency during boreal summer. The main injection of the shorter-lived tracers DMS and CH$_3$I remains over the tropical WIO, because their lifetimes are too short for injection through the summer monsoon circulation above northern India. Their transport efficiency also decreases in boreal summer due to weakened convection and vertical uplift above the WIO in comparison with spring. The annual cycle of stratospheric injection reveals the largest relative amplitude for DMS and the smallest amplitude for CH$_3$Br$_2$. This implies that the shorter the lifetime of a tracer, the stronger is the influence of the seasonal displacement of the main convection area and the strength of convection over the release area on stratospheric source gas injection.

Over the 16 years of our time series from January 2000 to February 2016, we found an increase in VSLS tracer transport efficiency from the WIO to the stratosphere for all tracers and during all seasons, which may be related to more intense vertical transport and a strengthening of the Asian monsoon circulation. Our transport efficiency using a rigid stratospheric threshold of 17 km is influenced by variations in the observed atmospheric tropopause height, and thus, the results may be impacted by the use of the tropopause as a threshold. The interannual variability of transport efficiency is highest for the shortest-lived tracer, DMS. Regarding individual seasons, interannual variability for all tracers is lowest during boreal spring, when the maximum transport occurs. The interannual variability of transport efficiency of all four VSLS tracers is influenced by the SST in the tropical WIO, as well as in the central equatorial Pacific. During boreal winter and spring, positive SST anomalies in the WIO and the Nino 4 region (El Niño) enhance stratospheric VSLS injection originating from the WIO. The warm Indian Ocean basin causes enhanced upward movement over the WIO, aiding the vertical transport to the stratosphere. During the summer monsoon, the transport of the two shortest-lived tracers for DMS and CH$_3$I is also influenced by high WIO SST anomalies and positive IOD events. In boreal fall, negative SST anomalies in the tropical Pacific (La Niña) cause stronger than normal stratospheric injection from the WIO through strengthening the monsoon flow from the Indian Ocean to the Indian subcontinent, and prolonging and bolstering the monsoon convection. A series of El Niño events may lead to more stratospheric injection above the WIO during boreal spring, while La Niña events enhance stratospheric injection of VSLS source gases above the central and northeast Indian Ocean during boreal fall.

In this study, we focus on variability and changes in the transport of VSLS from the West Indian Ocean to the stratosphere and do not account for the actual oceanic emissions. In a follow-up study, the temporal and spatial variability of oceanic VSLS emission of the whole tropical Indian Ocean will be considered. While we showed that the Asian monsoon circulation transports VSLS from the tropical West Indian Ocean to the stratosphere, we expect other subregions with high convective activity and hot spot emissions to be of even greater importance for VSLS delivery to the stratosphere.

Acknowledgments
This study was supported by BMBF grant SONNE-OASIS 03G0235A and the FP7 EU project StratoClim (603557). We thank the European Centre for Medium-Range Weather Forecasts (ECMWF) for the provision of ERA-Interim reanalysis data and the FLEXPART development team for the Lagrangian particle dispersion model used in this publication. Part of the FLEXPART simulations was performed on resources provided by UNINETT Sigma2—the National Infrastructure for High Performance Computing and Data Storage in Norway and with support of the IT staff of University of Oslo. WCRP and University of Oslo’s Industrial Liaison cooperation supported A. Fiehn’s participation at the Training School on Monsoon Variability in a Changing Climate in January 2017. We thank Roxy Mathew Roll for the helpful advice on influences on Indian monsoon variability. We would like to thank Yoshiyuki Kajikawa and Bin Wang for updating the Indian monsoon index on their website. C. A. Marandino is funded by the Helmholtz Young Investigator Group TRASE-EC (VH-NG-819) at the GEOMAR Helmholtz Centre for Ocean Research Kiel. The model output data used in this study are available at the NorStore Archive of UNINETT Sigma2 under the DOI: 10.11582/2016.00008. The authors declare that they have no conflict of interest.

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