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Interactive comment on "Delivery of halogenated very short-lived substances from the West Indian Ocean to the stratosphere during Asian summer monsoon" *by* Alina Fiehn et al.

Anonymous Referee #1

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Review for "Delivery of halogenated very short-lived substances from the West Indian Ocean to the stratosphere during the Asian summer monsoon" Alina Fiehn, et al. ACP-2017-8

Summary

The manuscript by Fiehn et al. presents observations of very short-lived species (VSLS) obtained from ship-based measurements during a cruise on the research vessel Sonne in the subtropical and tropical Indian Ocean in July and August 2014. The species observed are CH3I, CHBr3 and CH2Br2, all of which have potentially a strong impact on stratospheric chemistry and climate. Measurements of VSLS are sparse and show large variability. Attempts to create global emission estimates by creating

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Discussion paper

observation based climatologies or modeling are characterized by large uncertainties.

The observations presented consist of air and water samples and were obtained together with data from radio sondes launched during the cruise. Utilizing these local meteorological data compound specific transfer coefficients and subsequently air-sea fluxes of VSLS were calculated.

The paper does not only present observations, but includes transport simulations utilizing trajectories computed with the Lagrangian model Flexpart. The aim of this part of the study is on the identification of the source regions of the air masses observed as well as transport pathways of VSLS from the boundary layer into the stratosphere, followed by an estimation of the amount of the above mentioned species entrained into the stratosphere during the Asian summer monsoon. Different model set ups are used covering backward, forward and domain filling forward scenarios. In addition the study is applied to the results obtained during other measurement campaigns and extended to 16 years to specify inter-annual variability.

The results and conclusions presented show two major pathways of transport of VSLS from the West Indian Ocean boundary layer into the stratosphere, namely local convection on a time scale of 0-2 days and upward transport inside the Asian monsoon circulation over Nortehern India and the Bay of Bengal on a time scale of 6 to 13 days.

General

In general the manuscript is an organized and methodical sound paper. Starting from an introduction that embeds the studied processes into the current scientific background regarding the importance of biogenic VSLS for the stratospheric bromine and iodine budget as well as their role in stratospheric chemistry, and after having presented their observations, the authors focus on the role of vertical transport for the entrainment of VSLS into the stratosphere, the related problem of transit times and life times for different compounds and major pathways.

Aiming at these issues, the authors utilize the Flexpart trajectory model, a state-of-the-art transport model often used for investigations of transport processes in the troposphere. The model was driven by ERA Interim meteorological data with a 1 x 1 degree horizontal resolution. In first simple setups (OASIS backward and forward), the origin of the air masses probed along the ship track and their future trajectories are investigated. The OASIS forward trajectories already indicate the existence of different transport regimes, with upward transport within local convection over the West Indian Ocean and inside the Asian monsoon circulation above Northern India and the Bay of Bengal being the most prominent features. Entrainment into the stratosphere is supposed at 17 km height.

This leads to my first critical remark: The authors claim, that the value of 17 km height for the location of the tropopause represents the average cold point tropopause during the cruise and for the whole tropical region. This may well be true, but as one can depict from the figure S2, there is quite strong variability around the mean value, especially the more to the north one gets. Furthermore the authors show with quite a lot of effort results for different other heights to substantiate the choose of this value (section 5 and figure S3). But why not using a tropopause location computed from the ERA Interim data, the same data underlying the transport calculation? I am not doubting the general location of the maximum entrainment areas, but using the ERA Interim based tropopause would be much more convincing and more consistent.

Speaking of this, ERA Interim provides much higher horizontal resolution than used. Why not using it for better, more accurate trajectory results, better resolved convection and better tropopause location? Even the Flexpart parametrizations for vertical transport and convection could benefit from this.

To investigate more on stratospheric entrainment and the role of the West Indian Ocean a third forward trajectory model run is started with domain filling trajectories from a rectangular area over the area of interest (Indian Ocean setup). In this context now VSLS tracers are used, which undergo an exponential decay according to certain tropical tropospheric life times. This is in contrast to the vertical life time profile used for the simple OASIS setups, and I am not sure, why there are two different life times used. May be the authors can comment on this.

Nevertheless the results from this run and the results gained for different other campaigns and regional areas are quite interesting and plausible. The analysis of transit times, transport efficiencies and transit half times provides a good insight into the interplay of rapid local convection and slower upward motion (as inside monsoon circulations) and the impact on species with life times being much smaller than or comparable to the transport time.

There is one more remark about the investigtions with respect to the spatial varability of the stratospheric entrainment: In line 502 the definition of two core entrainment areas are mentioned, which are assumed to be evenly sized (and are shown in Fig. 6). It should be noted that these two regions may be evenly sized in grid space (that is 20 x 25 degrees), but not in area. Furthermore just by looking at the plots, one could think of moving the core entrainment box for the local convection 5 degrees more to the east for a better capture of entrainment. But maybe these boxes are chosen to be similar to Chen et al.(2012). If this is the case, it should be mentioned.

In the last part of the study the simulation with the Indian Ocean setup is extended to 16 years to specify inter-annual variability. To quantify the influence of different transport regimes (local convection and Asian monsoon) on the total entrainment, the respective time series are correlated by using Pearsons correlation coefficient. It should be noted, that for all values not -1 or 1 Pearsons r ist not meaningful, as long as there is no linearity between the two times series and/or if the values are not normally distributed. If there is a linear relationship, than correlation coefficients of 0.54 and 0.56 only explain 29% and 32% of the observed variance, meaning that roughly 70% of the variance is not explained. Even for a value of 0.87 (as for CH3I) just 75% of the variance are explained. To decide, whether these values are significant, you need to do a t test. A much more robust method, which does not imply linearity, but only monotonicity, is Spearmans rank correlation coefficient. There is a similar method introduced by Kendall. I would recommend to use one of these rank correlation coefficients, which is fairly easy to do, but would give more meaningful results.

Within the concluding section the main results are summarized clearly, followed by implications of the papers findings for global emission estimates for short-lived species and the influence of different transport pathways on the stratospheric entrainment of short-lived species. The authors discuss some potential problems regarding the calculation of vertical transport and uncertainties in the determination of the stratospheric entrainment with respect to the average cold point tropopause. Again, I would like to stress the point of calculating the local tropopause height directly from the used ERA Interim data, preferably from data with higher horizontal resolution.

Summarizing, the paper is well-written and presents an important contribution to our understanding of transport of the short-lived species from the boundary layer over the West Indian Ocean into the stratosphere. It should be published after some minor revisions.

Additional comments

The values shown in figure 6 are labeled as tracer density (given in percent). Is this meant to be the same as tracer entrainment?

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