"**The contribution of oceanic methyl iodide to stratospheric iodine**" by S. Tegtmeier et al.

Response to Anonymous Referee #1

We thank the reviewer for the positive feedback and helpful comments. We have addressed all reviewer comments in a revised manuscript. Our detailed answers to the comments can be found below in italics.

General remarks

In this manuscript Tegtmeier et al. revisit the question of the contribution of the short-lived volatile organic halogen CH3I to stratospheric iodine. By combining ship-based measurements of sea water concentrations, airborne measurements in the upper troposphere (& tropopause), and model results they present a detailed analysis of CH3I transport from the ocean surface into the upper tropical tropopause. Their results show that there are indeed instances of coinciding high oceanic emissions, high vertical transport, and high upper air mixing ratios that could imply temporary considerable contributions of the substance to stratospheric iodine. Overall, their detailed assessment of large-scale (from a global emission estimate & global Lagrangian modeling) and localized (from individual field campaigns) vertical transport of CH3I reveals that the contribution of this substance to stratospheric iodine has not yet been fully understood. As the paper shows interesting results and will trigger further investigations, I suggest publication in ACP. Below you find some minor basically technical remarks.

Detailed remarks

p11429, l16–18
.. where it plays an important role for .. the formation of ultrafine aerosol particles … Actually, the cited Saiz-Lopez et al. (2012) report that it is now believed that molecular iodine is the precursor of iodine-mediated ultrafine particles, rather than organic iodine-containing compounds. So CH3I's role in IOP formation is a little overstated.

The statement regarding the formation of ultrafine aerosol particles has been taken out.

p11429 l21–26
.. some studies report algae and phytoplankton as biological sources .. most studies suggest photochemical .. The differentiation between "some" and "most" is neither clear not trivial, here. Does it refer to the pure number of publications? It is simple to add two more that find CH3I production from macro- or micro-algae (e.g. Giese ea, ES&T 33, 2432-2439, 1999, Nightingale ea, Limnol. Oceanogr.40,680–689,1995) to even out the 6 mentioned that support the idea of photochemical production. Or does it refer to the number of independent (i.e. of different research groups) findings (clearly not, as two are inspired by D. Wallace and two by S. Manley)? Or does it indirectly refer to the (global?) relevance of the source? W.r.t. to the latter, is also not clear how to weight publications that use different methods to conclude about the source (co-variations, laboratory studies, modeling) when counting the number of articles. Generally, it should be noted that there is evidence for both production pathways. Their individual importance may vary with time, location, and water depth, and also for different research foci (e.g. halogen emissions, halogen cycling in sea water, processes determining specific observed concentrations).

Thanks for pointing this out. We have rephrased the sentence in order to not weigh the different mechanisms.

p11432 l16–19

.. fluxes were calculated from … sea surface concentration and .. mixing ratio applying Henry's law constant .. and the 10min average wind speed. Henry is a function of temperature. What temperature data did you use (also 10min ave.s?)?

Henry's law constant was calculated as a function of the 10-min average water temperature. The information has been added to the manuscript.

p11432 l20

The flux calculations are based on .. adapted to CH3I. Please be more specific on how this adaptation looks like (Schmidt number parametrization f(salinity,temperature)?). As the emissions are crucial for the findings it is important to know exactly what assumptions go into their calculation.

The text has been extended to ''The flux calculations are based on the transfer coefficient parameterization of Nightingale et al. (2000) adapted to CH_3I. For the parameterization, the transfer velocity at Schmidt number 660, which corresponds to CO_2 at 20°C in seawater (Wanninkhof, 1992), was corrected by the CH_3I Schmidt number at the temperature of measurement. The ratio of the diffusion coefficients from methyl bromide (De Bruyn and Saltzman, 1997) and methyl iodide, estimated according to Wilke and Chang (1955) was used as a function of temperature for the Schmidt-number correction (e.g., Richter and Wallace, 2004).''

p11433 l9–11

All campaigns took place over the S U.S. and C America. Over land? If so, are these obs. Still representative for the marine environment, i.e. can they be compared to the data obtained during ship cruises?

For the comparison we determine coincidences in time and space between the output of the global model runs and the aircraft measurements. Therefore, for each coincidence, the measurement and the model result should either both be representative of the land environment or both be representative of the maritime environment. The latter is mostly the case since the majority of the measurements were taken over the ocean or coastal regions (see map of flight tracks in Figure 6).

It is correct, that the here used global emissions maps do not include terrestrial emissions and therefore the model results over land could be an underestimation. However, a sensitivity study, where only coincidences over the ocean or coastal regions are included in the comparison, gives very similar results to the ones presented in the manuscript. This is very likely related to the fact that terrestrial emissions are expected to be considerably smaller than oceanic emissions (e.g., Barkley et al., 2007; Lai et al., 2011) and that most of the coincidences are found in the maritime environment. Therefore, we decided to keep the comparison as it is.

p11435 l9-11

A pre-processor retrieves the meteo. fields .. How relevant is this information?

The main information of the sentence is the fact that the vertical wind is calculated in hybrid coordinates mass consistently from spectral data and not that a pre-processor retrieves the meteorological fields. We have rephrased the sentence to make this clear.

p11436 l21–23

Emissions are based on an empirical parametrization of the gas transfer velocity that is not very well tested for high wind speeds. Please discuss the validity of the parametrization for/and its respective impact on emissions that occur at wind speeds higher than 20 ms1.

Indeed, it is true that the parameterization is based on low to moderate wind speeds. Existing different parameterizations provide a wide range of transfer coefficients and their dependency on the wind speed with the Nightingale parameterization approximately in the middle range. While there is discussion that the parameterized transfer coefficient at higher wind speeds could be an overestimation of the real transfer coefficient (e.g., McNeil and D'Asaro, 2007) the various processes determining the air-sea gas exchange at higher wind speeds (i.e., direct transfer associated with the air–sea interface, ventilation associated with surface wave breaking, bubble injection) are not well understood momentarily. Therefore, a thorough discussion of the validity of the parameterization at wind speeds higher than 20 ms^{-1} is beyond the scope of this manuscript and an ongoing research topic (e.g., Carpenter et al, 2012).

We agree with the reviewer that a short comment regarding the parameterization at higher wind speeds is necessary and have inserted the following text in the manuscript ''Among the different existing parameterizations, the here applied sea-to-air flux parameterization of Nightingale et al. (2000) predicts transfer velocities in the middle range (e.g., Carpenter et al., 2012) for wind speeds below 20 ms^{-1}. All parameterizations gain uncertainty for wind speeds above 20 ms^{-1} and a possible overestimation of sea-to-air fluxes at these very high wind speeds has been suggested (McNeil and D'Asaro, 2007). While this needs to be kept in mind when fluxes at higher wind speeds are considered, for the here discussed cruises wind speeds are always below 20 ms^{-1} and the Nightingale parameterization has been applied throughout.''.

p11436 l23

Reference to Quack et al.2013: articles in preparation are unfort.ly not directly available to the reader.

We apologize for not making a draft version of this manuscript available to the reviewers. If Quack et al. 2013 (momentarily in preparation) will not be available through ACPD by the time our manuscript is accepted we will remove the citation.

p11457 Tab.1

According to the definition mentioned on p11430 Bell et al. (2002) use a “bottom-up” approach (based on [CH3I] obtained during ship cruises) to derive emissions. They calculate aqueous [CH3I] in a slab ocean model and thereby constrain their production rates by a least-squares fit of model results to observed sea water concentrations derived during ship cruises. Emissions are than calculated from the standard two-film model, and mostly influenced by sea water concentrations, as the ocean is strongly over-saturated.

Thanks for pointing this out. We have changed the table entry.

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