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Interactive comment on “The contribution of oceanic methyl iodide to stratospheric iodine” by S. Tegtmeier et al.

Anonymous Referee #1

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General remarks

In this manuscript Tegtmeier et al. revisit the question of the contribution of the short-lived volatile organic halogen CH_3I 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 CH_3I 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 CH_3I reveals that

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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.

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).

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p11432 I16–19 .. 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?)?

p11432 I20 The flux calculations are based on .. adapted to CH_3I
Please be more specific on how this adaptation looks like (Schmidt number parametrization $f(\text{salinity}, \text{temperature})?$). As the emissions are crucial for the findings it is important to know exactly what assumptions go into their calculation.

p11433 I9–11 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?

p11435 I9–11 A pre-processor retrieves the meteo. fields ..
How relevant is this information?

p11436 I21–23 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 ms^{-1} .

p11436 I23 Reference to Quack et al.2013: articles in preparation are unfort.ly not directly available to the reader.

p11457 Tab.1 According to the definition mentioned on p11430 Bell et al. (2002) use a “bottom-up” approach (based on $[\text{CH}_3\text{I}]$ obtained during ship cruises) to derive emissions. They calculate aqueous $[\text{CH}_3\text{I}]$ 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

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the standard two-film model, and mostly influenced by sea water concentrations, as the ocean is strongly over-saturated.

References

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- Nightingale, P. D., Malin, G., Law, C., Watson, A., Liss, P., Liddicoat, M., Boutin, J., and Upstill-Goddard, R.: In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers, *Global Biogeochem. Cycles*, 14, 373–387, 2000.
- Saiz-Lopez, A., Plane, J. M. C., Baker, A. R., Carpenter, L. J., von Glasow, R., Martín, J. C. G., McFiggans, G., and Saunders, R. W.: Atmospheric Chemistry of Iodine, *Chemical Reviews*, 112, 1773–1804, 10.1021/cr200029u, 2012.

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