Is halogen chemistry responsible for low ozone levels observed over western Pacific equatorial region? - From observations on R/V Mirai

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Constraints from ozone (O_3) observations over oceans are needed in addition to those from terrestrial regions to fully understand global tropospheric chemistry and its impact on the climate. Here, we provide a large data set of ozone and carbon monoxide (CO) levels observed (for 11666 and 10681 h, respectively) over oceans. The data set is derived from observations made during 24 research cruise legs of R/V *Mirai* during 2012 to 2017, in the Southern, Indian, Pacific, and Arctic Oceans, covering the region from 67˚S to 75˚N. The data are suitable for critical evaluation of the over-ocean distribution of ozone derived from chemical transport models. We first give an overview of the statistics in the data set and highlight key features in terms of geographical distribution and air mass type. We then use the data set to evaluate ozone concentration fields from Tropospheric Chemistry Reanalysis version 2 (TCR-2), produced by assimilating a suite of satellite observations of multiple species into a chemical transport model, namely CHASER. Over the western Pacific equatorial region (125-165˚E, 10˚S to 25˚N), the observed O_3 level frequently decreased to less than 10 ppb in comparison to that obtained with TCR-2, and also those obtained in most of the Atmospheric Chemistry Climate Model Intercomparison Project (ACCMIP) model runs for the decade from 2000. These results imply loss processes that are unaccounted for in the models. We found that the model's positive bias positively correlated with the daytime residence times of air masses over a particular grid, namely 165-180˚E and 15-30˚N; an additional loss rate of 0.25 ppb h^{-1} in the grid best explained the gap. Halogen chemistry, which is commonly omitted from currently used models, might be active in this region and could have contributed to additional losses. Our open data set covering wide ocean regions is complementary to the Tropospheric Ozone Assessment Report data set, which basically comprises ground-based observations, and enables a fully global study of the behavior of O_3.

Keywords: Over-ocean atmospheric composition observations, Missing sink of ozone, Halogens, Western Pacific equatorial region

