Impacts of tropospheric bromine and iodine on global tropospheric ozone: a modeling study using CHASER

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Bromine and iodine catalytic cycles are recognized as additional and important sinks of tropospheric ozone, though most of global climate transport models do not consider processes relevant to tropospheric bromine and iodine compounds (Young et al., 2018). A few global models have been used to evaluate impacts of tropospheric bromine and iodine on global tropospheric ozone (e.g., Saiz-Lopez et al., 2014; Sherwen et al., 2016), while large uncertainties in emission model process representation and emission estimation still remain. We newly implement iodine chemistry process, short-lived halocarbon emission, and ozone-mediated inorganic iodine release from the ocean (Chang et al., 2004; Carpenter et al., 2013) in the CHASER chemical transport model (Sudo et al., 2002; Sekiya et al., 2018) and evaluated impacts of tropospheric bromine and iodine on global tropospheric ozone. Incorporating these processes reduced surface ozone concentration by 20% on the annual and global average over the ocean. The model which incorporates these processes showed better agreements with ship-borne in-situ observations on the R/V Mirai and Hakuho-maru (Kanaya et al., 2019) during 2014-2018 (mean bias = 0.04 ppbv, $r = 0.82$) than the standard model (mean bias = 2.36 ppbv, $r = 0.73$). We evaluated impacts of tropospheric bromine on global tropospheric ozone loss using different three oceanic emission estimates of short-lived brominated halocarbons ($CHBr_3$, CH_2Br_2, CH_2BrCl, $CHBr_2Cl$, and $CHBrCl_2$): a bottom-up estimate based on SeaWIFS satellite observations of chlorophyll-a (Ordóñez et al., 2012), a bottom-up estimate for $CHBr_3$ and CH_2Br_2 using ship-borne observations (Ziska et al., 2013), and a top-down estimate for $CHBr_3$, CH_2Br_2, $CHBr_2Cl$, and $CHBrCl_2$ derived from the ATOM aircraft-campaign observations (Wofsy et al., 2018) using the Bayesian inversion technique. When using these three emissions, the global tropospheric ozone loss owing to bromine were estimated to be 284, 72, and 211 Tg O_3/yr, respectively, which suggested that bromine-catalyzed loss highly depends on short-lived halocarbon emissions. For tropospheric iodine sources, oceanic iodine release (as HOI and I_2) triggered by heterogeneous ozone loss at the sea surface was dominant over iodinated halocarbons (CH_3I, CH_2ICl, CH_2IBr, and CH_2I_2). The global tropospheric ozone loss owing to atmospheric iodine chemistry was estimated to be 643 Tg O_3/yr, which was comparable to the values reported by previous studies. Furthermore, model results suggested that the initial heterogeneous ozone loss at the sea surface (192 Tg O_3/yr) was an important sink of global tropospheric ozone which has not been evaluated explicitly in the previous studies. These results demonstrated the importance of the processes related to tropospheric bromine and iodine on global tropospheric ozone. In future studies, it is necessary to get a better understanding of tropospheric bromine and iodine compounds' emission processes and to reduce their uncertainties.

Keywords: Tropospheric ozone, Halogen, Chemical transport model

