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Vertical and spatial distribution of Chloromethane and Bromomethane from boundary layer to upper troposphere over the Amazon rainforest

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Chloromethane (CH_3Cl, average ambient mixing ratio 546 ppt (1)) and Bromomethane (CH_3Br, average ambient mixing ratio 6.52 ppt (1)) are the most abundant chlorine-, respective bromine-containing atmospheric trace gases from natural origin. Due to their relatively long tropospheric lifetimes (0.9 y; 0.8 y respectively (2)), they are significant carriers of chlorine and bromine into the stratosphere, where they are photolyzed and contribute to the catalytic destruction of ozone. Chloromethane is mainly emitted by tropical vegetation and soils, biomass burning and from oceans. Vegetation and soils have been shown to act as both, source and sink for Chloromethane. Bromomethane has anthropogenic and biogenic sources, including oceans, fumigation, biomass and fossil fuel burning, crops and vegetation. For both methyl halides the known tropospheric sinks, such as reaction with hydroxyl radicals, loss to soils and oceans, and loss to the stratosphere, do not balance the currently known sources. In particular the strengths of the tropical sources still have substantial uncertainties. Due to the Montreal Protocol, anthropogenic emissions of chlorine and bromine compounds are declining. Therefore, biogenic contributions will become an even more important fraction of the global budget in the following decades. A better understanding of the mechanisms behind emission and tropospheric distribution of Chloro- and Bromomethane is essential to improve predictions of future stratospheric ozone levels.

Here, we present the first airborne measurements of Chloromethane and Bromomethane over a tropical forest covering altitudes from the planetary boundary layer to the upper troposphere (300 – 14000 m). The measurements were conducted in Dec 2022 and Jan 2023 over the Amazon Rainforest with the German research aircraft HALO (High Altitude Long Range) in the scope of the CAFE-Brazil campaign. Chloromethane, Bromomethane and other Halocarbons and VOC were measured with Fast GC-MS with a time resolution of 3 minutes. On average elevated levels of both species were found in the boundary layer. Interestingly, the vertical distribution of both compounds also showed a layer with elevated mixing ratios in the upper troposphere, generating a C-shaped profile. The mean Chloromethane mixing ratio reached a maximum of 590 ± 30 ppt between 11 - 12 km altitude, whereas for Bromomethane the maximum appears to be higher than 14 km (where 11.8 ± 1.9 ppt were observed) and thus outside the covered altitude range. The high local convective activity and that of the more distant ITCZ may explain these observations. Surprisingly, high variability of mixing ratios in the boundary layer (300 – 1000 m) with seasonal

and regional trends for both methyl halides was observed. Chloromethane mixing ratios between 499 and 686 ppt were observed, in agreement with earlier works which report that the rainforest (including vegetation and soil) can act as source and sink of Chloromethane. Bromomethane mixing ratios in the boundary layer varied between 6.0 to 20.9 ppt, indicating the rainforest to be a source of Bromomethane.

(1) World Meteorological Organization (WMO). Scientific Assessment of Ozone Depletion: 2022. Geneva; 2022. Report No.: 278.

(2) World Meteorological Organization (WMO). Scientific Assessment of Ozone Depletion: 2018. Geneva; 2018. Report No.: 58.