Toward a High Degree of Freedom Full Atmosphere Retrieval of BrO Profiles from MAX-DOAS Instruments on Remote Tropical Marine Mountaintops

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The remote tropical troposphere is responsible for about 75% of the chemical removal of ozone (O_3) and methane (CH_4); two important greenhouse gases. Yet the atmospheric chemistry of tropospheric halogens over remote oceans is largely unconstrained in the free troposphere (FT) where natural background processes can be probed in absence of local impacts from pollution. Inorganic bromine and iodine radicals from ocean sources are responsible for about 20% of the global tropospheric ozone loss, equivalent to about 900 Tg O_x yr^{-1} (similar to the O_x loss from hydroperoxyl). Halogens oxidize atmospheric mercury, modify aerosols, and iodine can form new particles. The Volkamer group at the University of Colorado Boulder is developing a small network of mountaintop Multiple AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) instruments to probe hemispheric gradients in the remote tropical FT by long-term measurements of trace gases. Since February 2017 we have deployed MAX-DOAS instruments at two sites: 1) Mauna Loa Atmospheric Baseline Observatory (MLO) at 19.5° N, 155.6° W, at 3.4 km altitude in the northern hemisphere tropics, and 2) Maïdo Observatory (Maïdo) at 21.1° S, 55.4° E, at 2.2 km altitude in the southern hemisphere tropics. We measure the halogen oxide radicals bromine oxide (BrO) and iodine oxide (IO), small oxygenated volatile organic compounds (OVOC; e.g. formaldehyde and glyoxal), as well as total columns of O_3, and nitrogen dioxide (NO_2), and aerosol optical depth, which can be used for satellite validation. Leveraging O_3 and NO_2 columns, and coupling photochemical change into radiative transfer we retrieve stratospheric profiles from twilight zenith measurements with additional degrees of freedom. Actively determining the reference contribution extends the degrees of freedom in tropospheric profile retrievals. Corroborating previous studies we find comparable total columns of BrO in the troposphere and stratosphere. In the troposphere we consistently find low concentrations in the lower troposphere contrasting with higher concentrations in the upper FT. We examine the implications of this trace gas distribution for development of high degree of freedom retrievals of vertical columns and atmospheric profiles aiming at up to 5 degrees of freedom.

Figure 1. A) MODIS imagery of La Réunion Island in the southwest Indian Ocean from June 11, 2017. Arcs indicate the viewing direction of the Royal Belgian Institute for Space Aeronomy (BIRA) operated MAX-DOAS in Le Port on the coast and the CU-Boulder MAX-DOAS at Maïdo Observatory at 2.16 km. **B)** The Maïdo Observatory. The MAX-DOAS telescope is mounted on the roof with the rest of the instrument below it. During the intensive phase other instruments will be on the roof, or use inlets extending above the roof, or horizontally from the observatory. **C)** The CU-Boulder MAX-DOAS telescope. The telescope can move in the vertical plane perpendicular to photo to gather profile information by probing different altitudes.