

Figure S1. *Top panel:* Maps of total vertical column density of BrO measured on 17 April 2008 by OMI (left) and GOME-2 (right). *Middle panel:* Estimates of stratospheric vertical column density of BrO (Strat BrO) found using the formulation adopted in the main body of our paper (UMCP; see main text) (left) and found using the formulation of Theys et al. (2011) (right), all for OMI overpass time. *Bottom panel:* Map of tropopause pressure at time of OMI overpass from MERRA (see main text) (left) and the ratio of Strat BrO from UMCP divided by Strat BrO from Theys formulation (right). The ratio of these two formulations of Strat BrO bears a relation to tropopause pressure, because the UMCP-based estimate has a higher contribution to total column from air in the lowermost stratosphere (LMS) than the Theys-based estimate, due to the greater emphasis of Product Gas Injection of bromine in the UMCP

formulation. When air in the LMS is compressed to high density (high tropopause pressure), the difference in the two formulations grows. Variations in solar zenith angle with respect to latitude and height profiles of total inorganic bromine as a function of altitude also factor into the ratio. For the day shown, the mean value of the ratio of these two estimates of Strat BrO is 1.37. For the 8 days for which data appear in the main paper (5, 6, 7, 8, 17, 19, 21, and 22 April), the mean and standard deviation of the value of the ratio is 0.27 ± 0.09 .

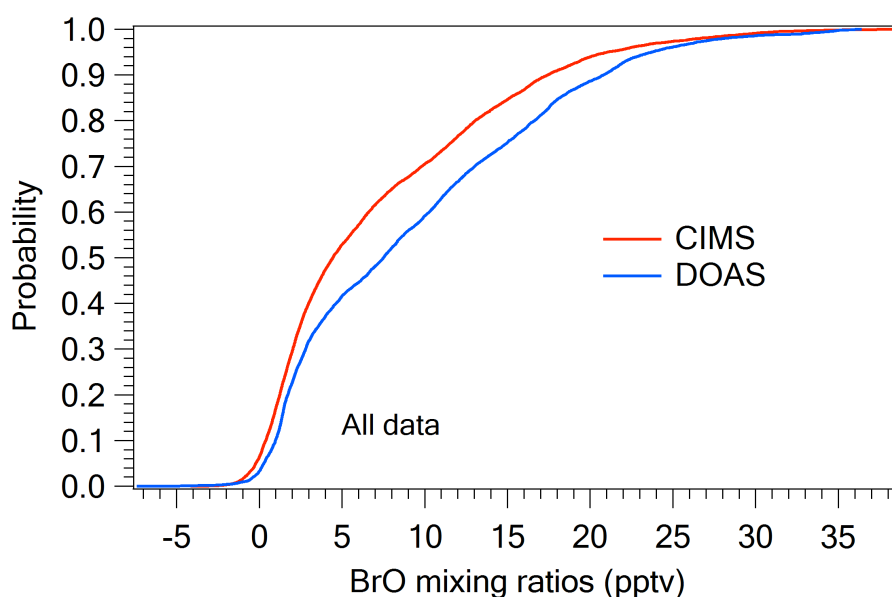


Figure S2. Probability Distribution Function (PDF) of surface BrO measured by two ground-based instruments at Barrow, Alaska in spring 2009: a chemical ionization mass spectrometer (CIMS) and a long path-differential optical absorption spectrometer (LP-DOAS). Only data acquired during daytime are represented. This PDF corresponds to the data shown in Figure 5 of Liao et al., JGR, doi:10.1029/2010JD014788, 2011. While surface BrO measured by CIMS and LP-DOAS peaked at 41 and 42 pptv, respectively, the PDF shows that 2/3 of the time, BrO at the surface as measured by CIMS was below 8 pptv. This determination is important for the main body of the paper due to the following: for a surface temperature of 245 K, density at the surface would be $\sim 3 \times 10^{19}$ molecules/cm³. The lowest altitude sampled by the CIMS instrument on board the DC-8, for descents in regions BrO-enhanced regions, was 75 meter above the surface. In the main body of the paper, we use a composite DC-8 profile to estimate the in-situ tropospheric column. Suppose the surface BrO measurements of Liao et al. (2011) were present, uniformly, between the surface and 75 meters. Then the resulting column, for 8 pptv of BrO, would be 0.18×10^{13} molecules/cm² ($8 \times 10^{-12} \times 3 \times 10^{19}$ molecules/cm³ \times 7500 cm), which is a small contribution to the total in-situ column as well as the total vertical column of BrO. The PDF shown in Figure S2 supports the view that most of the time, the perturbation to our analysis due to an enhanced layer of BrO present below the aircraft would be small and inconsequential. If 40 pptv of BrO had been uniformly distributed between the surface and 75 meter altitude would contribute $\sim 1 \times 10^{13}$ molecules/cm² to these columns, which would be consequential. The PDF shows 40 pptv of BrO, while seen, was rarely present. As a result, we state in the main paper “while layers of highly elevated BrO below the aircraft could on occasion compromise our comparisons, our overall conclusions are robust because surface measurements indicate only on rare occasion are BrO enhancements large enough to significantly perturb the column.”