REMOTE SENSING OF BROMINE MONOXIDE IN RELATION TO BOUNDARY LAYER HALOGEN CHEMISTRY

W. R. Simpson[1], D. Donohoue[1], D. Carlson[1], R. Salawitch[2], T. Canty[2], T. Kurosu[3], K. Chance[3]

[1]Department of Chemistry and Geophysical Institute, University of Alaska, Fairbanks, USA

[2]Atmospheric and Oceanic Sciences, University of Maryland, College Park, Maryland, USA

[3]Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA

Remote sensing of bromine monoxide (BrO) from the Ozone Monitoring Instrument (OMI) and other satellite instruments opens the possibility to extend our understanding of atmospheric halogen chemistry from local-scale studies to global scales. In the Arctic springtime boundary layer, these bromine monoxide radicals and bromine atoms are known to destroy ozone [1] and oxidize mercury [2], leading to its deposition and possible incorporation into the food chain. In the stratosphere, these same reactive bromine species deplete ozone, affecting the ground-level UV dose. The satellite BrO vertical column density (VCD) data contain contributions from both boundary layer BrO and stratospheric BrO. Therefore, it is critical to separate the stratospheric BrO from the boundary layer BrO to determine in which atmospheric layer the satellite-detected BrO is causing impacts. In this presentation, we describe ground-based remote sensing of boundary layer BrO and combine these observations with satellite observations and models to develop and test methods for partitioning satellite-detected total column BrO measurements.

Reactive halogens (*e.g.* BrO) are produced in the boundary layer through gas-surface reactions on the surface of ice that contains sea salts [1]. While the exact mechanism of what types of salt-containing surfaces are most responsible for reactive halogen production is a matter of debate, the process is clearly related to the salinity of ice surfaces. The salinity of sea ice surfaces during the Arctic springtime is increasing due to dramatic reductions in perennial (multi-year) sea ice in the summer, which increases the coverage of the more saline first-year sea ice. Therefore, it is critical to measure the boundary layer component of BrO so as to be able to relate its abundance to sea ice properties. With a firm understanding of the relationship between halogen activation and sea ice properties, it should be possible to make informed predictions regarding the impacts of summer sea ice reductions on boundary layer ozone depletion and

mercury deposition. Satellite observations of total column BrO VCDs show sporadic "hotspots" of halogen activation primarily over the frozen Arctic Ocean along with lower levels ubiquitously around the Arctic. Traditionally, we have interpreted these data by assuming the stratospheric background leads to the ubiquitous background levels and the hotspots are tropospheric (boundary layer) events. In this presentation, we use ground-based remote sensing observations of BL BrO from Barrow, Alaska to attribute the satellite-detected total column BrO to BL and non-BL (primarily stratospheric) BrO. Three models of non-BL BrO are tested. The first is a constant stratospheric background, the second is a model where non-BL BrO is correlated to total column ozone, and the third is a stratospheric chemical model produced by Salawitch and Canty. From this testing, we find that the total column BrO from OMI observes boundary layer BrO events and that the constant stratospheric background model is insufficient to explain the data and variable non-BL BrO is required for column closure. The chemical model gives the best results, although the second method gives nearly as good results. This result indicates that quantitative extraction of BL BrO from OMI requires accurate modeling of the non-BL component.

At other regions around the Arctic, we see persistent hotspots in the OMI total column BrO observations. A particularly prominent hotspot is in the Hudson Bay area, which has been observed since the first Global Ozone Monitoring Experiment (GOME) BrO VCD observations [3]. Due to lack of ground-based observations of BrO, like we used at Barrow, Alaska, we cannot do a complete column closure analysis to partition this satellite-detected BrO hotspot into BL and non-BL components. However, analysis of the minimum BrO/O_3 ratio at these locations appears to indicate that a larger fraction of the BrO "hotspot" over Hudson Bay is not in the boundary layer as compared to the Barrow non-BL component.

This work has major implications for using satellite BrO observations to study boundary layer BrO events. From the Barrow observations, we have shown that a variable non-BL (primarily stratospheric) BrO abundance is necessary to extract BL BrO information. The analysis in the Hudson Bay area indicates that area has a different non-BL BrO abundance than Barrow, indicating that subtraction of non-BL BrO is spatially variable. It is critical to both further develop methods for removal of non-BL BrO and observe BL BrO at varying locations around the globe (*e.g.* Hudson Bay) to test these methods. The presence of a variable and larger

than expected lower-stratospheric BrO burden indicated by the chemical model of Salawitch and Canty may also have significant implications for stratospheric ozone depletion.

Bibliography

[1] Simpson, W. R., von Glasow, R., Riedel, K., Anderson, P., Ariya, P., Bottenheim, J., Burrows, J., Carpenter, L. J., Frieß, U., Goodsite, M. E., Heard, D., Hutterli, M., Jacobi, H.-W., Kaleschke, L., Neff, B., Plane, J., Platt, U., Richter, A., Roscoe, H., Sander, R., Shepson, P., Sodeau, J., Steffen, A., Wagner, T., and Wolff, E.: Halogens and their role in polar boundary layer ozone depletion, Atmos. Chem. Phys., 7, 4375-4418, 2007.

[2] Steffen, A., Douglas, T., Amyot, M., Ariya, P., Aspmo, K., Berg, T., Bottenheim, J., Brooks, S., Cobbett, F., Dastoor, A., Dommergue, A., Ebinghaus, R., Ferrari, C., Gardfeldt, K., Goodsite, M. E., Lean, D., Poulain, A. J., Scherz, C., Skov, H., Sommar, J., and Temme, C.: A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow, Atmos. Chem. Phys., 8, 1445-1482, 2008.

[3] Richter, A., Wittrock, F., Eisinger, M., Burrows, J. P.: GOME Observations of Tropospheric BrO in Northern Hemisphere Spring and Summer of 1997, Geophys. Res. Lett., 24(14), 2683-2686, 1998.