Volumes and Issues | Contents of Issue 22

Atmos. Chem. Phys., 9, 8651-8660, 2009
www.atmos-chem-phys.net/9/8651/2009/

Sensitivity of polar stratospheric ozone loss to uncertainties in chemical reaction kinetics

S. R. Kawa[1], R. S. Stolarski[1], P. A. Newman[1], A. R. Douglass[1], M. Rex[2], D. J. Hofmann[3], M. L. Santee[4], and K. Frieler[2,*]

[1]NASA Goddard Space Flight Center, Greenbelt, MD, USA
[2]Alfred Wegener Institute for Polar and Marine Research, Potsdam, Germany
[3]National Oceanic and Atmospheric Administration, Earth Systems Research Laboratory, Boulder, CO, USA
[4]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA
[*]now at: Potsdam Institute for Climate Impact Research (PIK), Potsdam, Germany

Abstract. The impact and significance of uncertainties in model calculations of stratospheric ozone loss resulting from known uncertainty in chemical kinetics parameters is evaluated in trajectory chemistry simulations for the Antarctic and Arctic polar vortices. The uncertainty in modeled ozone loss is derived from Monte Carlo scenario simulations varying the kinetic (reaction and photolysis rate) parameters within their estimated uncertainty bounds. Simulations of a typical winter/spring Antarctic vortex scenario and Match scenarios in the Arctic produce large uncertainty in ozone loss rates and integrated seasonal loss. The simulations clearly indicate that the dominant source of model uncertainty in polar ozone loss is uncertainty in the Cl_2O_2 photolysis reaction, which arises from uncertainty in laboratory-measured molecular cross sections at atmospherically important wavelengths. This estimated uncertainty in $J_{Cl_2O_2}$ from laboratory measurements seriously hinders our ability to model polar ozone loss within useful quantitative error limits. Atmospheric observations, however, suggest that the Cl_2O_2 photolysis uncertainty may be less than that derived from the lab data. Comparisons to Match, South Pole ozonesonde, and Aura Microwave Limb Sounder (MLS) data all show that the nominal recommended rate simulations agree with data within uncertainties when the Cl_2O_2 photolysis error is reduced by a factor of two, in line with previous in situ ClO_x measurements. Comparisons to simulations using recent cross sections from Pope et al. (2007) are outside the constrained error bounds in each case. Other reactions producing significant sensitivity in polar ozone loss include BrO + ClO and its branching ratios. These uncertainties challenge our confidence in modeling polar ozone depletion and projecting future changes in response to changing halogen emissions and climate. Further laboratory, theoretical, and possibly atmospheric studies are needed.

Final Revised Paper (PDF, 1192 KB)   Discussion Paper (ACPD)

Citation: Kawa, S. R., Stolarski, R. S., Newman, P. A., Douglass, A. R., Rex, M., Hofmann, D. J., Santee, M. L., and Frieler, K.: Sensitivity of polar stratospheric ozone loss to uncertainties in chemical reaction kinetics, Atmos. Chem. Phys., 9, 8651-8660, 2009.   Bibtex   EndNote   Reference Manager