Introduction and Abstract

Several recent observational and laboratory studies of processes involved in polar stratospheric ozone loss have prompted a reappraisal of our understanding of this key indicator of global change. To a large extent, our confidence in understanding and projecting changes in polar and global ozone is based on our ability to simulate these processes in numerical models of chemistry and transport. The fidelity of the models is assessed in comparison with a wide range of observations. These models depend on laboratory-measured kinetic reaction rates and photolysis cross sections to simulate molecular interactions. The rates of all of these reactions are subject to uncertainty, some substantial. In particular, recent lab measurements of the ClO + ClO photolysis cross sections (Pope et al., 2007) are significantly different (smaller) than those reported in the latest JPL rate compilation (Sander et al., 2006).

In this study we use a simple box-model scenario for Antarctic ozone to estimate the uncertainty in loss attributable to known reaction kinetic uncertainties. Following the method of earlier work (Stolarski et al., 1978, Stolarski and Douglass, 1986), rates and uncertainties from the latest laboratory evaluation are applied in random combinations. We determine the key reactions and rates contributing the largest potential errors and compare the results to observations to evaluate which combinations are consistent with atmospheric data. Implications for our theoretical and practical understanding of polar ozone loss are highlighted.

ANTARCTIC OZONE

Rate Sensitivity

**Key Reaction Uncertainties**

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Rate File</th>
<th>+1σ</th>
<th>-1σ</th>
</tr>
</thead>
<tbody>
<tr>
<td>ClO + ClO + M</td>
<td>JPL'06</td>
<td>0.125</td>
<td>-0.125</td>
</tr>
<tr>
<td>Br + OClO</td>
<td>JPL'06</td>
<td>0.666</td>
<td>-0.666</td>
</tr>
<tr>
<td>BrCl + O</td>
<td>Pope et al.</td>
<td>0.75</td>
<td>-0.75</td>
</tr>
<tr>
<td>HBr + HCO</td>
<td>JPL'06</td>
<td>0.5</td>
<td>-0.5</td>
</tr>
</tbody>
</table>

Summary

- Known uncertainties in kinetic reaction rate parameters from laboratory measurements produce significant uncertainty in Antarctic O3 loss calculated in a simple, but representative, model.
- The impact of varying ClO2 cross sections between JPL'06 and Pope et al. is distinguishable at the 95% confidence level in a spring Antarctic O3 loss scenario.
- Comparison to observations shows the ozone sonde and MLS data are consistent with JPL'06 rates but not Pope et al. within model uncertainty.
- Both data sets suggest somewhat faster O3 loss needed in the model relative to the base case.
- Findings are consistent with previous work with earlier rate compilations at mid latitudes and in the Arctic (Fah and Burton [1997], Rex et al.).

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References:


**Model Uncertainty Scenarios**

- Large range of calculated O3 loss within JPL'06 error limits
- Significantly slower loss with Pope et al. ClO2 cross sections

**Ozone Loss Statistics**

- ClO2 cross-section scenarios are distinct at the 95% confidence level.

**COMPARISON WITH OBSERVATIONS**

- Measured O3 in each year favors JPL'06 scenario or even faster loss.
- MLS and models ClO increase consistent with respective O3 loss.

**Sensitivity of Polar Stratospheric Ozone Loss to Uncertainties in Chemical Reaction Kinetics**

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