Contribution ID: **9** Type: **Poster**

The Effect of the Novel HNO3(g) Production

Monday, 6 June 2011 12:05 (1h 10m)

It is well established that the reaction of HO2 with NO plays a central role in atmospheric chemistry by way of OH/HO2 recycling and reduction of ozone depletion by HOx cycles in the stratosphere and in ozone production in the troposphere. In the stratosphere this reaction moderates the effectiveness of the cycle involving HOx radicals, which is an important removal mechanism of ozone. In the troposphere this reaction plays a key role in controlling the interconversion between HO2 and OH radicals through cycles involving CO and volatile organic compounds (VOCs); these chemical cycles serve as a secondary source of OH radicals as well as a major source of tropospheric ozone.
Utilizing a photochemical box model, we investigate the impact of the recently observed HNO3 production channel (HO2 + NO ⟶ HNO3) on NOx (NO + NO2), HOx (OH + HO2), HNO3, and O3 concentrations in the boundary layer at South Pole, Antarctica. The inclusion of the new reaction channel decreases peak O3, NO, NO2, and OH by 3 ppbv, 200 pptv, 125 pptv, and 0.3 pptv, respectively, from their peak concentrations, while peak HNO3 increases by 125 pptv; the peak HO2 amount is unaffected by the new chemistry. The decrease in O3 from 37 to 34 ppbv worsens the agreement between the model and the highest O3 concentrations observed at the South Pole (45 ppbv). As the mean concentration of OH at the South Pole is 2.5 × 106 molecules cm-3, our simulations brings mean OH into better agreement with observations as OH is reduced from 3 × 107 to 1 × 107 molecules cm-3. HO2 are incongruent with maximum and mean concentrations of HO2 measured at the South Pole, ˜ 7 × 107 and ˜ 1 × 106 molecules cm-3, respectively. Given that peak measured HNO3 concentration measured at the South Pole is 70 pptv, our simulations still over-predict it by an additional ˜ 130 pptv. The reduced concentrations of NOx as a result of the application of the new channel are consistent with expected decreases in atmospheric NOx lifetime as a result of increased sequestration of NOx into HNO3. Without this new channel current models will tend to underestimate NOx losses. This has important implications: 1) given that previous studies investigating snowpack emissions of NOx have relied upon accurate and comprehensive determinations of NOx losses in this environment, our result implies that NOx snowpack emissions are larger than currently reported; 2) current estimates of the nitrate recycling factor again rely upon accurate characterization of NOx lifetime, and, therefore, current estimates of the nitrate recycling factor are likely too low. Although we show that the inclusion of the novel HNO3 production channel brings better agreement for HOx with field measurements, the modeled ozone and HNO3 are worsened, and the changes in NOx lifetime imply that snowpack NOx emissions and snowpack nitrate recycling must be re-evaluated.

Please list some keywords

snowpack, sea-ice, ice photochemistry

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Session Classification: Lunch and Poster