Observations of HO_x and its relationship with NO_x in the upper troposphere during SONEX

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Abstract

Simultaneous measurements of the oxides of hydrogen and nitrogen made during the NASA Subsonic Assessment, Ozone and Nitrogen Oxide Experiment (SONEX) afforded an opportiunity to study the coupling between these two important families throughout the free troposphere and lowermost stratosphere. Moreover, the suite of measurements made during the campaign was unprecendented in its completeness, thus providing a uniquely detailed picture of the radical photochemistry that drives oxidation and ozone production in this part of the atmosphere. On average, observed hydrogen oxides (HO_x = OH + HO_2) agree well with both instantaneous and diel steady state models; however, there is a persistent deviation of the observations that correlates with the abundance of nitrogen oxides (NO_x = NO + NO_2) in the sampled air mass. Specifically, the observed HO_x tends to exceed the model predictions in the presence of high NO_x concentrations, by as much as a factor of 5 (> 500 pptv NO_x) and is sometimes as little as half that expected by steady state at lower NO_x levels. While many possibilities for these discrepancies are discussed, it is argued that an instrumental artifact is not probable and that the discrepancy may bespeak a shortcoming of our understanding of HO_x chemistry. The consistently elevated HO_x in the presence of elevated NO_x leads directly to greater ozone production than expected, thereby extending the NO_x-limited regime of the upper troposphere. These results could thus have bearing on the predicted impacts of increasing NO_x emissions into this region of the atmosphere from, for example, the growth of global air traffic.
