

The Impact of Current CH4 and N2O Loss Process Uncertainties on Model Calculated Ozone and Global Lifetimes

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The atmospheric loss processes of CH4 and N2O, their estimated uncertainties, lifetimes, and impacts on ozone abundance and long-term trends are examined using atmospheric model calculations and updated kinetic and photochemical parameters and uncertainty factors from SPARC (2013). The largest stratospheric impacts are due to uncertainty in the O(1D)+N2O reaction, both in the total rate coefficient and branching ratio for the O₂+N2 and 2*NO product channels. This uncertainty results in a substantial range in calculated present day stratospheric odd nitrogen ($\pm 10\text{--}25\%$) and global total ozone ($\pm 1\text{--}2.5\%$). The O(1D)+N2O reaction uncertainty also affects the rate of past global total ozone decline and future recovery, with a range in future ozone projections of $\pm 1\text{--}1.5\%$ by 2100, relative to present day. Uncertainty in the Cl+CH4 reaction affects the amount of chlorine in radical vs. reservoir forms and has a modest impact on present day SH polar ozone ($\sim \pm 6\%$), and on the rate of past SH polar ozone decline and future recovery. Uncertainties in the photolysis of N2O and the CH4 loss due to reaction with OH and O(1D) have relatively small impacts on present day calculated global total ozone ($\pm 0.2\text{--}0.4\%$), with the OH+CH4 uncertainty impacting tropospheric ozone by $\pm 3\text{--}5\%$. The ranges in calculated CH4 and N2O global lifetimes due to the kinetic and photochemical uncertainties are also examined: these ranges are significantly reduced when using the updated SPARC estimated uncertainties compared with those from JPL-2010. As a possible application of this work, the reduced uncertainty in the lifetimes can be used to help refine projections of future N2O and CH4 concentrations.