

Chemical NO_x budget in the upper troposphere over the tropical South Pacific

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Abstract

The chemical NO_x budget in the upper troposphere over the tropical South Pacific is analyzed using aircraft measurements made at 6-12 km altitude in September 1996 during the Global Tropospheric Experiment (GTE) Pacific Exploratory Mission (PEM) Tropics A campaign. Chemical loss and production rates of NO_x along the aircraft flight tracks are calculated with a photochemical model constrained by observations. Calculations using a standard chemical mechanism show a large missing source for NO_x; chemical loss exceeds chemical production by a factor of 2.4 on average. Similar or greater NO_x budget imbalances have been reported in analyses of data from previous field studies. Ammonium aerosol concentrations in PEM-Tropics A generally exceeded sulfate on a charge equivalent basis, and relative humidities were low (median 25% relative to ice). This implies that the aerosol could be dry in which case N₂O₅ hydrolysis would be suppressed as a sink for NO_x. Suppression of N₂O₅ hydrolysis, together with adoption of new measurements of the reaction rate constants for NO₂+OH+M and HNO₃+OH reduces the median chemical imbalance in the NO_x budget for PEM-Tropics A from 2.4 to 1.9. The remaining imbalance cannot be easily explained from known chemistry or long-range transport of primary NO_x, and may imply a major gap in our understanding of the chemical cycling of NO_x in the free troposphere.

The full text of this paper is available as a [postscript file](#)