FINAL REPORT

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ABSTRACT

Our work has two primary scientific goals, the interannual variability (IAV) of stratospheric ozone and the hydrological cycle of the upper troposphere and lower stratosphere. Our efforts are aimed at integrating new information obtained by spacecraft and aircraft measurements to achieve a better understanding of the chemical and dynamical processes that are needed for realistic evaluations of human impact on the global environment. A primary motivation for studying the ozone layer is to separate the anthropogenic perturbations of the ozone layer from natural variability. Using the recently available merged ozone data (MOD), we have carried out an empirical orthogonal function (EOF) study of the temporal and spatial patterns of the IAV of total column ozone in the tropics. The outstanding problem about water in the stratosphere is its secular increase in the last few decades. The Caltech/JPL multi-dimensional chemical transport model (CTM) photochemical model is used to simulate the processes that control the water vapor and its isotopic composition in the stratosphere. Datasets we will use for comparison with model results include those obtained by the Total Ozone Mapping Spectrometer (TOMS), the Solar Backscatter Ultraviolet (SBUV and SBUV/2), Stratosphere Aerosol and Gas Experiment (SAGE I and II), the Halogen Occultation Experiment (HALOE), the Atmospheric Trace Molecular Spectroscopy (ATMOS) and those soon to be obtained by the Cirrus Regional Study of Tropical Anvils and Cirrus Layers Florida Area Cirrus Experiment (CRYSTAL-FACE) mission. The focus of the investigations is the exchange between the stratosphere and the troposphere, and between the troposphere and the biosphere.

ACCOMPLISHMENTS

Temporal and spatial patterns of the interannual variability of total ozone in the tropics

The recently constructed dataset, combining the monthly mean column abundances collected by the Total Ozone Mapping Spectrometer (TOMS) and the Solar Backscatter Ultraviolet (SBUV and SBUV/2) instruments, provides a nearly continuous record from late 1978 to 2000 on a $5^\circ \times 10^\circ$ latitude-longitude grid. The precision and calibration of these measurements allow very small signals, ~1% of total column ozone, to be clearly seen. Using merged ozone data (MOD), we have carried out an empirical orthogonal function (EOF) study of the temporal and spatial patterns of the interannual variability of total column ozone in the tropics. The first four EOFs of our study capture over 94% of the variance of the deseasonalized data. The leading two EOFs, respectively accounting for 54% and 25% of the variance, display structures attributable to the quasi-biennial oscillation (QBO), with influence from a decadal oscillation (most likely the solar cycle). The third EOF (11% of the variance) represents an interaction between the QBO and an annual cycle. The fourth EOF (4% of the variance) is related to the El Nino - Southern Oscillation. This is the first complete analysis of tropical ozone data using simultaneously the longitudinal, latitudinal and temporal patterns. A paper based on this work (Camp *et al.* 2003) is published in *J. Geophys. Research*. The preprint may be viewed in http://yly-mac.gps.caltech.edu/ under "O_3 Paper".

Isotope Compositions in the Upper Troposphere and Lower Stratosphere

Observations of the isotopic ratio in H_2O vapor can provide unique tests of the atmospheric physics that control dehydration. With improved H_2O linelists and retrieval methods, we are able to estimate the water isotope abundances in the upper troposphere/lower stratosphere, especially for the tropics, using Fourier Transform InfraRed (FTIR) Interferometer data from the Atmospheric Trace Molecule Spectroscopy (ATMOS) and the Mark IV balloon flights. The D/H ratio remains constant from 10 to 25 km (left panel), even though the mixing ratio of H_2O changes by an order of magnitude (right panel). This provides definitive evidence for the tropical tropopause layer (TTL), first suggested by Sherwood and Dessler (2001). The details of this work may be viewed from our recent AGU presentation (Kuang *et al.*, 2001; http://www.gps.caltech.edu/~kzm/HDO/). Simultaneous retrieval of CO from the same spectra yields vertical profiles for this short-lived molecule. From this an estimate of the time constant of mixing in the TTL can be made. A paper has appeared in Geophys. Res. Lett.

Publications related to this project

C.E. Miller and Y.L. Yung, 2000, Photo-induced isotopic fractionation, *J. Geophys. Res.*, **105** 29039-29051.

C.D. Camp, M.S. Roulston, A.F.C. Haldemann, and Y.L. Yung, 2001, The sensitivity of tropospheric methane to the interannual variability in stratospheric ozone, *Chemosphere-Global Change Science,* **3,** 147-156.

Kuang, Z. M., J. S. Margolis, G. C. Toon, D. Crisp, and Y. L. Yung, 2002, Spaceborne measurements of atmospheric CO_2 by high-resolution NIR spectrometry of reflected sunlight: A sensitivity Study, Submitted to *Geophys. Res. Lett.* **29** (15), 11-1, 11-4.

C. D. Camp, M. S. Roulston, and Y. L. Yung, 2003, Temporal and spatial patterns of the interannual variability of total ozone in the tropics, *J. Geophys. Res.*, **108** (D20), 4643, doi:10.1029/2001/JD001504.

Z. M. Kuang, G. C. Toon, P. O. Wennberg and Y. L. Yung, 2003, Measured HDO across the tropical tropopause, *Geophys. Res. Lett.*, **30**(7): art. no. 1372.

T. K. Tromp, R. L. Shia, M. Allen, and Y. L. Yung, 2003, Potential environmental impact of a hydrogen economy on the stratosphere, *Science* **300**, 1740-1742.

Huang, X, and Yung, Y.L., 2004. A common misunderstanding about the Voigt line profile. *J Atmos. Sci.* **61**, 1630-1632.

Morgan, C. G., Allen, M., Liang, M. C., Shia, R. L., Blake, G. A. and Yung, Y.L., 2004. Isotopic fractionation of nitrous oxide in the stratosphere: Comparison between model and observations. *J Geophys. Res.* **109** (D4) art. No. D04305.

Conference Presentations

Mills, F. P., Sander, S. P., Cageao, R. P., Nemtchinov, V., Jiang, Y., Russell, J. M., Remsberg, E. E., LeBlanc, T., LeBlanc, T., Nedoluha, G., Allen, M. A., and Yung, Y. L., Intercomparisons of OH Column Measurements from the JPL Table Mountain Facility with Measurements of O_3 and H_2O, EOS Transactions American Geophysical Union, Vol. 81, No. 48, A71B-13, p.F99, December 15-19, 2000.

E. J. Moyer, et al., 2001, Seasonal Variation in Episodic Rapid Meridional Transport in the Lowermost Stratosphere from in-situ Trace Gas Measurements, EOS Transactions American Geophysical Union, Vol. 81, No. 48, A42B-20.

C.G. Morgan, G.A. Blake, T. Rahn, and Y.L. Yung, 2001, Isotopic Fractionation of N_2O in the Stratosphere Revisited, Paper presented at the 18th Informal Conference on Photochemical and Kinetics Process in the Atmosphere, February 20, California Institute of Technology, Pasadena, CA.

Z. M. Kuang, et al., 2001, Isotope Compositions in the Upper Troposphere and Inside the Polar Vortex, EOS Transactions American Geophysical Union, Vol. 82, No. 47, A12C-10.

J. Farrara, X. Jiang, S. S. Leroy, J. Feynman, A. Ruzmaikin, and Y. Yung, 2001, Effects of a Reduced Ozone Layer on the Lower Stratosphere and the Troposphere, AGU Fall Meeting, A11C-06, San Francisco, CA.

Camp, C. D., Sheetz, N. A., and Yung, Y. L., "Presence of a Madden-Julian Oscillation Signal in the TOMS Ozone Data." Atmospheric Sciences [A], 2002, A52A-0100 AGU Fall Meeting

Yung, Y. L., "The mean isotopic composition of nitrous oxide inferred from atmospheric modeling." Biogeosciences [B], 2002, B71A-0700 AGU Fall Meeting

Liang, M. C., C. D. Parkinson, A. Y. Lee, Y. L. Yung and S. Seager, 2003, A Comparative Study of Photochemistry in the Atmospheres of an Extrasolar. EOS Transactions American Geophysical Union, Vol. 84, No. 46, A12A-0074, p.113, Dec 8-12, 2003.

Noone, D., J. Randerson and Y. L. Yung, 2003, The effect of Arctic warming and sea ice loss on the growing season in northern terrestrial ecosystems. EOS Transactions American Geophysical Union, Vol. 84, No. 46, B12G-02, p.118, Dec 8-12, 2003.

Jiang, X., C. D. Camp, R. L. Shia, D. Noone, C. Walker, T. Schneider and Y. L. Yung, 2003, QBO and QBO-annual Beat Signals in the Tropical Total Column Ozone Simulated by a Two-dimensional Chemistry and Transport Model. EOS Transactions American Geophysical Union, Vol. 84, No. 46, A21D-1015, p.142, Dec 8-12, 2003.

Huang, X., Y. L. Yung and B. H. Lambrigtsen, 2003, Spatial-Spectral EOF analysis of AIRS data: an exploratory study. EOS Transactions American Geophysical Union, Vol. 84, No. 46, H32B-0569, p.236, Dec 8-12, 2003.

Camp, C. D., J. Feynman, X. Jiang, R. L. Shia, C. Walker, T. Schneider, M. Allen and Y. L. Yung, 2003, Solar Cycle Variation in the Ozone Distribution Simulated by a Two-dimensional Chemistry Transport Model. EOS Transactions American Geophysical Union, Vol. 84, No. 46, SH52B-06, p.353, Dec 8-12, 2003.

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E. J. Moyer, Tracers of Rapid Transport Processes in the Lower Stratosphere, 2001, California Institute of Technology.

Z. M. Kuang, I: Remote Spectroscopic Measurements of Atmospheric HDO/H_2O and Column CO_2, II: Interannual Variations of the Earth's Reflectance, 2003, California Institute of Technology.