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A. Stratospheric Data Analysis System (STRATAN)
(673-41-43-20)

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B. The goal of this project is to produce state-of-the-art stratospheric analyses using a coupled stratosphere-troposphere data assimilation system (STRATAN). These analyses can be applied to stratospheric studies of all types. Of importance to this effort is the application of STRATAN to constituent transport and chemistry problems (Stratospheric General Circulation with Chemistry Model (SGCCM), RTOP # 673-64-01-20)

C. During the past two years, 3 months of assimilation have been completed from the 1978-1979 Limb Infrared Monitor of the Stratosphere (LIMS) period. Assimilation for January and February 1989 have been completed. The 1989 experiments were in support of an Airborne Arctic Stratosphere Expedition theory proposal (R. Stolarski, PI). Both of these assimilation experiments are continuing.

STRATAN analyses have been compared to NMC (National Meteorology Center) analyses and to atmospheric profile data. These comparisons indicate that STRATAN does indeed produce a state-of-the-art analysis. These comparisons have also indicated several problem areas that compel improvements to the assimilation system.

The most important product of STRATAN is the wind analysis. The winds from STRATAN are very different from geostrophic winds from the NMC analyses and compare much better to direct wind measurements. Transport calculations confirm that winds from STRATAN are indeed a better measure of stratospheric winds.

The quality of the winds from STRATAN demonstrate the use of assimilation techniques in stratospheric analysis. We plan to initiate constituent assimilations in the next proposal cycle. Assimilation of data into comprehensive dynamical and photochemical models provide s a mechanism for taking full advantage of all the space and time information that is contained in satellite and ground based measurements.

E. The publication list showing the most recent publications for the Stratospheric General Circulation with Chemistry Project is attached. Numerous papers are in preparation.

SGCCP PUBLICATIONS (as of 9/13/89)

Published Papers

32. Rood, R. B., D. J. Allen, W. E. Baker, D. J. Lamich, and J. A. Kaye, "The use of assimilated stratospheric data in constituent transport calculations," J. Atmos. Sci., 46, 687-701, 1989.
38. Wu, M. F., M. A. Geller, E. R. Nash, and M. E. Gelman, "Global atmospheric circulation statistics--four year averages," NASA Technical Memorandum 100690, June 1987.
41. Geller, M. A., and M.-F. Wu, "Troposphere-stratosphere general circulation statistics," Transport Processes in the Middle Atmosphere, (G. Visconti and R. Garcia, eds.), D. Reidel Publishing Co., 3-17, 1987.
42. Chao, W. C., "On the origin of the tropical intraseasonal oscillation," J. Atmos. Sci., 44, 1940-1949, 1987.
43. Kaye, J. A., and R. B. Rood, "Chemistry and transport in a three-dimensional stratospheric model chlorine species during a simulated stratospheric warming," J. Geophys. Res., 94, 1057-1083, 1989.
44. Rosenfield, J. E., M. R. Schoeberl, and P. A. Newman, "Antarctic spring-time ozone depletion computed from temperature observations," J. Geophys. Res., 93, 3833-3849, 1988.
45. Kaye, J. A., "Mechanisms and observations for isotope fractionation of molecular species in planetary atmospheres," Rev. Geophys., 25, 1609-1658 1987.
46. Geller, M. A., M.-F. Wu, and E. R. Nash, "Satellite data analysis of ozone differences in the Northern and Southern Hemispheres," Pure & Appl. Geophys., 130, 263-275, 1989.

47. Geller, M. A., "Solar cycles and the atmosphere," Nature, 332, 584-585, 1988.
48. Kaye, J. A., R. B. Rood, and D. J. Allen, "Variability of chlorine containing species as revealed by three-dimensional stratospheric transport and chemistry models," Proceedings of Quadrennial Ozone Symposium, August 8-13, 1988, Gottingen, FRG.
49. Rood, R. B., and J. A. Kaye, "Stratospheric ozone models and super-computers," Proceedings of the Fourth International Conference on Supercomputing, April 30-May 5, 1989, Santa Clara, California.
50. Rosenfield, J. E., "A simple parameterization of ozone infrared absorption for atmospheric heating rate calculations," J. Geophys. Res., (submitted), 1989.
51. Rood, R. B. J. A. Kaye, D. J. Allen, W. E. Baker, and D. J. Lamich, "The use of winds and temperatures from a stratospheric assimilation model in three-dimensional constituent transport studies," Proceedings of Quadrennial Ozone Symposium, August 8-13, 1988, Gottingen, FRG.
52. Rood, R. B., J. A. Kaye, A. R. Douglass, D. J. Allen, S. Steenrod, and E. M. Larson, "Wintertime nitric acid chemistry: Implications from three-dimensional model calculations," J. Atmos. Sci., (submitted), 1989
53. Kaye, J. A. and R. B. Rood, "Simulations of Short-Term Variability of Stratospheric Trace Constituents," Proceedings of the 28th International Astrophysical Colloquium, "Our Changing Atmosphere," Leige, Belgium, June 1989.
54. Kaye, J. A., R. B. Rood, D. J. Allen, E. M. Larson, and C. H. Jackman, "Three Dimensional Simulation of Spatial and Temporal Variability of Stratospheric Hydrogen Chloride," Geophys. Res. Lett., (accepted), 1989.

Interpretation of Satellite and In Situ Aerosol and Trace Gas Data using Chemical-Dynamical Models.

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Objectives: We utilize global 3D chemical-dynamical-radiative models, analytical dispersion models, and statistical theory to address: (a) interpretation of recent satellite and in situ data on stratospheric aerosols which play important roles in the global energy budget and in the stratospheric sulfur cycle; (b) interpretation of satellite and in situ data on several trace gases of importance to our understanding of the chemistry of the stratospheric ozone layer; and (c) better definition of the spatial and temporal resolution required for determination of ozone trends from a network of ozone-sonde or lidar stations. The satellite data of interest come from NIMBUS-7 (SAMS, SAM II, LIMS), SAGE-1 and SME. The in situ data come from balloons, aircraft, and ground-based observations (including the Global Atmospheric Gases Experiment - GAGE).

Summary of 1988-1989 progress:

A critical examination of ozone variability and instrumental precision indicates that the current ozonesonde network is insufficient to detect a trend in tropospheric ozone of $\leq 1\%$ per year at the 2σ level even at stations with records a decade in length. We have concluded that in order to detect a 1% per year trend in a decade or less it will be necessary to decrease the time between observations from its present value of 3-7 days to 1 day or less. We have developed an analytical theory taking into account photochemistry, surface deposition, and wind climatology to define the "effectively sampled region" for an observing station which forms the basis for defining a suitable global network for determining regional and global ozone trends; a paper describing the work has been published (Prinn, 1988a).

We have also completed a detailed review of atmospheric halocarbons (CF_2Cl_2 , CH_3Cl , CCl_3F , CH_3CCl_3 , CCl_4 , CHClF_2 , CF_4 , $\text{C}_2\text{Cl}_3\text{F}_3$, CH_3Br , CBrClF_2 , CH_3I) and products of halocarbon decomposition (HCl , HF , ClO , ClONO_2) with the goal of identifying major shortcomings and/or contradictions in the available data. Largest trends are seen for CHClF_2 and $\text{C}_2\text{F}_3\text{Cl}_3$ but absolute calibration and global emission estimates for these 2 species are very uncertain. Also data on the stratospheric trends in HCl , ClO , and ClONO_2 are uncertain or nonexistent and the trends that have been reported for HF are not consistent with trends in their fluorocarbon sources. This work has also been published (Prinn, 1988b).

The annual percentage increases in concentrations of the chlorofluorocarbons $\text{CF}_2\text{ClCFCl}_2$ (CFC-113, an industrial solvent) and CHClF_2 (CFC-22, a refrigerant) are the highest among major chlorofluorocarbons in the atmosphere today. We have computed the present-day atmospheric lifetimes for these species using our global three-dimensional dynamical-chemical model. The present-day lifetimes of both are long (15.5 years for CHClF_2 , and 136 or 195 years for $\text{CF}_2\text{ClCFCl}_2$ depending on assumed O_2 absorption cross-sections) underscoring the need to decrease their emissions in order to minimize their future role in ozone destruction and greenhouse warming. These results have been published (Golombek and Prinn, 1989a).

The compound 1,1-dichloro-2,2,2-trifluoroethane (CHCl_2CF_3 ; CFC-123) is a proposed replacement for trichlorofluoromethane (CFCl_3 ; CFC-11). We have computed the chemical destruction rates by various mechanisms and the steady-state lifetime of CFC-123 using our global three-dimensional chemical-dynamical model. The major destruction mechanism for this species is reaction with the hydroxyl radical in the troposphere below the altitude of 12 km which accounts for 89% of its total loss rate. The computed steady-state lifetime for CFC-123 is 1.87 ± 0.07 years (1σ) which is very much shorter than that for CFC-11 which is destroyed largely in the stratosphere. For equal mass emission rates of CFC-123 and CFC-11 into the atmosphere, the steady-state atmospheric content and steady-state injection rate of chlorine into the stratosphere are respectively 35 and 15 times less for CFC-123 than for CFC-11. A paper on this work by Prinn and Golombek has been submitted to *Nature*.

We have also studied the processes maintaining the non-volcanically-perturbed stratospheric sulfuric acid (Junge) layer using our global three-dimensional model. The model includes production of SO_2 from OCS, oxidation of SO_2 to gaseous H_2SO_4 , condensation-evaporation equilibrium of gaseous and particulate H_2SO_4 , condensation growth of particulates as they enter the tropopause-upper troposphere region, and particle rainout in the lower troposphere. We have compared our results with the NIMBUS 7 SAM II and AEM-2 SAGE stratospheric aerosol extinction data for periods when the stratosphere was not perturbed by recent volcanic eruptions. The model simulates the general behavior of stratospheric aerosol extinction including the existence of a polar tropopause enhancement in this extinction. Agreement is good in the tropics but there is a tendency however for the model in high latitudes to overpredict aerosol extinction above 15 km due perhaps to an overly vigorous predicted circulation or to inadequate knowledge of particle sizes. We identify two major non-volcanic sources for stratospheric H_2SO_4 : one is upwardly transported and photodissociated OCS and the other is upwardly transported SO_2 . The importance of SO_2 is a new and significant result. A paper on this work by Golombek and Prinn has been submitted to the *Journal of Atmospheric Chemistry*.

Publications:

1. Prinn, R. (1988a). Toward an improved global network for determination of tropospheric ozone climatology and trends. *J. Atmos. Chem.*, **6**, 281-298.
2. Prinn, R. (1988b). How have the atmospheric concentrations of the halocarbons changed? In "The changing Atmosphere" (ed. S. Rowland and I. Isaksen, J. Wiley and Sons Ltd.) pp. 33-48.
3. Golombek, A., and R. Prinn (1989). Global 3-dimensional model calculations for the budgets and present-day atmospheric lifetimes of $\text{CF}_2\text{ClCFCl}_2$ (CFC-113) and CHClF_2 (CFC-22). *Geophys. Res. Lett.*, in press for October 1989 issue.