

## A32B MC: Hall D Wednesday 1330h

### New Insights Into Stratospheric Chemistry, Dynamics, and Transport III

*Presiding:* E Shuckburgh, Ecole Normale Supérieure

#### A32B-0038 1330h POSTER

##### Small Scale Transports of Trace Gases in the Upper Troposphere/Lower Stratosphere as Related to Eddy Diffusivities

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Small scale fluctuation of trace gases in the UT/LS are discussed in terms of eddy transports. Mixing ratios of H<sub>2</sub>O, CFC11, HNO<sub>3</sub>, and O<sub>3</sub> are analyzed, that were measured by the CRISTA experiment. Fluctuations are seen to be dependent on latitude and altitude. The results are compared to the eddy diffusivities of Haynes and Shuckburgh (JGR, 2000). Trace gas transports are estimated. It is checked whether the atmosphere fluctuation field is homogeneous and stationary.

#### A32B-0039 1330h POSTER

##### Impact of mixing on the ozone chemistry in late Arctic spring 2000: Simulations with the Chemical Lagrangian Model of the Stratosphere (CLaMS)

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High resolution simulations of the chemical composition of the Arctic stratosphere during late spring 2000 are made with the Chemical Lagrangian Model of the Stratosphere (CLaMS). The simulations are performed for the entire northern hemisphere on four isentropic levels (400-475 K) until the last vortex remnants are irreversibly mixed at the beginning of June with the mid-latitude air masses. The initialization in early February is based on observations made from satellite, balloon, and ER-2 aircraft platforms.

The mixing intensity in CLaMS is controlled by the finite-time Lyapunov exponent  $\lambda$  measuring the deformation rate of the horizontal wind and switching on mixing in the flow regions where  $\lambda$  exceeds a critical value  $\lambda_c$ . A comparison of the CLaMS results with the tracer-tracer correlations observed *in situ* in early March and with HALOE CH<sub>4</sub> mixing ratios observed end of May in the vortex remnants indicates an inhomogeneous mixing in the lower stratosphere, both in time and space, if  $\lambda > \lambda_c = 1.2 \text{ day}^{-1}$ , with lateral (across the wind) effective diffusion of the order  $10^3 \text{ m}^2 \text{ s}^{-1}$ . Between early February and mid March, 2000, the Arctic vortex was well-isolated in the altitude range 400-475 K without significant mass exchange across the vortex edge. After the vortex breakup, slow dilution of the vortex remnants significantly influences the spatial distribution of ozone poleward of 30°N.

By varying the effective diffusion between 0 and  $10^4 \text{ m}^2 \text{ s}^{-1}$ , the impact of mixing on the ozone depletion is considered. In particular, the deactivation of ClO<sub>x</sub> within the air masses originating from the polar vortex occurs, even after the vortex breakup, mainly through the photochemical NO<sub>x</sub> production from HNO<sub>3</sub> without a significant contribution from mixing with NO<sub>x</sub>-rich mid-latitude air. The mid-latitude (30-60°N) ozone deficit in late boreal spring 2000 is dominated

by an irreversible transport of the ozone-depleted polar air masses (dilution) and, until mid of April, by ClO<sub>x</sub>-induced ozone destruction within the vortex remnants. Compared with the ozone distribution in a chlorine-free stratosphere, the mid-latitude ozone deficit at isentropic level 450 K amounts end of May to about 12% and is of the same order as in the high-latitudes (>60°N).

#### A32B-0040 1330h POSTER

##### STRATOSPHERIC OZONE VARIABILITY DURING THE POLAR SUMMER SEASON

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The transport and mixing of ozone and other chemically active trace constituents in the summer stratosphere has become the focus of recent studies. This interest is partly due to the failure of three-dimensional chemical transport models to fully reproduce the summer ozone distribution in the polar regions.

A new European-wide project funded by the EU Environment and Climate Programme "Spring-to-Autumn Measurements and Modelling of Ozone and Active Species" (SAMMOA) started in March 2000, with the goal to better understand the issues of ozone transport and chemistry in the summertime northern high latitudes. SAMMOA is coordinated by the Norwegian Institute for Air Research (NILU), and comprises eight other European research institutes. The project relies on using an integrated approach combining ground-based and balloon-borne measurements, global satellite observations, as well as advanced chemical-dynamical modelling and data assimilation.

In the frame of this project, summertime daylight ozone lidar measurements were made at the Arctic Lidar Observatory for Middle Atmosphere Research (Andoya, Norway) by NILU, the Norwegian Defence Research Establishment and the Andoya Rocket Range, as well as simultaneous total ozone measurements, both in 2000 and 2001. The 2000 total ozone record revealed the characteristic annual variation with a maximum in April and a minimum in September, which is mainly due to a variation in tropopause height. In addition, the total ozone showed a secondary oscillation with an amplitude of about 30 DU and a period of 25-30 days from mid April to mid-October. The lidar measurements combined with ozonesonde measurements from the closest sonde station, Sodankylä, indicate that this oscillation is caused by features in the middle and upper stratosphere ( $z > 25 \text{ km}$ ). The signature of the upper stratosphere patterns varies, however, considerably from year to year, and a preliminary evaluation of the 2001 data shows a much less pronounced oscillation with a larger period. Experimental data and corresponding model results will be presented.

#### A32B-0041 1330h POSTER

##### Studies of stratospheric and mesospheric chemistry in polar regions with the Berlin Climate Middle Atmosphere Model (CMAM CHEM)

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The Berlin Climate Middle Atmosphere model (CMAM) with resolution T21 and lid at 84km (Langematz and Pawson, 1997) has been interactively coupled with the chemistry module of Steil et al., (1998).

Chemical tracer fields from a test run which used the Palmer gravity wave drag (GWD) scheme have been found to compare well both with observations and other models (e.g., MA-ECHAM4-CHEM). However, in this run the model pole has a warm bias and dynamical variability is not well reproduced. Therefore we are currently testing a suite of orographic and non-orographic GWD schemes in the model. Preliminary results from these latter runs are presented here and focus on stratospheric and mesospheric chemical processes during polar winter, particularly those affecting ozone. The contrast between hemispheres, the springtime 'recovery' of polar stratospheric ozone and relevant chemical diurnal cycles are explored.

Bibliography

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B. Steil, Dameris M., Brhl C., Crutzen P. J., Grewe V., Ponater M., and Sausen R., Development of a chemistry module for GCMs: first results of a multiannual integration, Ann. Geophys., 16, 205-228, 1998

#### A32B-0042 1330h POSTER

##### Total Ozone Data From a European Network 1951-1957

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Soon after its foundation in 1948, the International Ozone Commission (IOC) established a total ozone network in Europe, together with the Gassiot Committee of the Royal Society, UNESCO, the London Meteorological Office and national services. The network was built-up in 1950 with Dobson spectrophotometers equipped with photomultipliers, which were calibrated in Oxford before shipping to the stations. In 1957, some of the stations became part of the network of the IGY, and these data can be found today at the WOUDC. The earlier data were compiled and archived in Oxford by the secretary of the IOC, Charles Normand, but have never been published and only rarely appeared in the scientific literature [Normand, QJRMS 67 (1951) 474 and QJRMS 69 (1953) 39]. The copies of the data sheets stored at UK Met Office [MO/19/3/9 Part I] comprise daily values from the following stations/time periods: Aarhus (DK, 6/52-12/59, Dobson #41), Aldergrove (UK, 6/52-4/57, #35?), Arosa (CH, 6/52-12/58 #15), Cagliari/Elmas (IT, 12/54-5/59, #48), Camborne (UK, 1/52-12/59, #32), Eskdalemuir (UK, 9/57-12/59, #35), Hemsby (UK, 6/52-9/55), Lerwick (UK, 6/52-12/59, #7), Magny les Hameaux (FR, 1/55-9/57, #49?), Messina (IT, 7/54-6/58, #46), Oxford (UK, 6/52-12/59, #1), Paris/Montsouris (FR, 10/57-8/58, #49), Reykjavik (IS, 6/52-10/59, #50), Rome/Vigna di Valle (IT, 4/54-12/59 #47), Santa Maria/Azores (ES, 2/53-7/56, #13), Spitzbergen (NO, 11/50-7/58, #8), Tromsø (NO, 6/52-5/59, #14), Uccle (BE, 6/52-12/58, #40), and Uppsala (SE, 6/52-12/58, #30).

These data could be useful to supplement the currently available total ozone measurement series. Together with existing meteorological data, they enable us to study the relation between atmospheric circulation and total ozone in a chemically largely unperturbed time period. The daily values from 1951 to 1957 have now been digitized. Using appropriate statistical methods, the quality of each series will be addressed. The data will be homogenized and re-examined in the framework of the project "Total ozone and climate Variability over Europe" funded by the Swiss NSF. An outline of the project is presented.

URL: <http://sinus.unibe.ch/~broenn/totalozon.page.htm>

#### A32B-0043 1330h POSTER

##### Effect of Quasi Stationary Wave 1 on Ozone Distribution During November 1993

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Microwave Limb Sounder (MLS) observations show southern hemisphere ozone and potential temperature fields that are dominated by quasi stationary wave number one during November 1993. These fields show wave 1 ozone and potential temperature perturbations that are in phase in the lower stratosphere (30 hPa) and in opposition of phase in the upper stratosphere (2 hPa), indicating the role of vertical transport induced by the wave in the presence of vertical gradient. Ozone and potential temperature gradients have the same sign in the lower stratosphere and opposite sign in the upper stratosphere. Ozone field reconstructed using temperature perturbation and the vertical mean gradient is comparable to the observations in the lower stratosphere; while in the upper stratosphere, the amplitude of the reconstructed ozone is smaller, indicating the presence of other process. Diabatic heating field calculated using MLS data and the NCAR radiation model exhibits in the upper stratosphere a strong wave 1 structure, which tend to damp temperature perturbations, inducing cross isentropic ozone transport. This result is supported by ozone distribution on isentropic surface that show a residual wave 1 fluctuation in the upper stratosphere, which is small in the lower stratosphere.

Ozone field evolution in the lower stratosphere computed using back trajectories technique shows large filament like equator-ward transport of ozone depleted air over South America region. This transport seems to be associated with the quasi stationary wave, which induce an upward displacement of isentropic surfaces in this region, reducing air density and the potential vorticity gradient.

### A32B-0044 1330h POSTER

#### Observations of Ozone Laminae From SAGE II (V 6.0) Measurements

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Laminar structures in the vertical profiles of ozone mixing ratio are commonly observed in ozonesonde measurements in the lower stratosphere in the middle and high latitudes. However there are a limited number of sounding stations and a global perspective can be obtained only from satellite data. Improved vertical resolution (< 1 km) has made it possible to resolve laminae in ozone mixing ratio profiles as retrieved by the Stratospheric Aerosol and Gas Experiment (SAGE II) version 6.0. A statistical study of these laminae in the latitude range 30-75 N and S and in the altitude range 10-28 km was carried out using 12 years (1985-1990, 1994-1999, excluding the years when the measurements were affected by the Pinatubo volcano) of SAGE II data. The occurrence peaks near 12-13 km, then decreases sharply up to about 20 km before increasing again in the 20-28 km range. The seasonal variation indicates that laminar activity maximizes during winter-spring in both hemispheres. This is consistent with earlier results from ozonesondes and satellites. Contrary to earlier climatologies, a significant number of laminae were observed in summer in both hemispheres primarily at high latitudes (> 60 N and S). These might represent remnants of air from polar vortex which undergo slow mixing for a long time after the vortex break up.

### A32B-0045 1330h POSTER

#### On the Vertical Alignment of an Atmospheric Vortex

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During its early stages of development, an atmospheric vortex can be destroyed by episodes of external vertical shear. However, some vortices survive, because they have a dominant tendency to stand upright.

We will review a recent theory for the vertical alignment of a quasi-geostrophic (QG) vortex.<sup>1</sup> In this theory, vertical alignment occurs by the damping of a discrete vortex Rossby mode. The damping rate  $\gamma$  (i.e., the alignment rate) is proportional to the potential vorticity gradient at a critical radius, where the rotation frequency of the vortex is resonant with the mode. Furthermore,  $\gamma$  varies with the vortex height and the level of atmospheric stratification. We find that  $\gamma$  can either increase or decrease with these parameters, depending on the radial structure of the vortex.

We will also present a more general theory that covers the vertical alignment of rapidly rotating vortices, such as tropical cyclones. This new theory is based on the asymmetric balance equations of Shapiro and Montgomery.<sup>2</sup> As in QG theory, vertical alignment is viewed as the damping of a discrete mode. However, the damping rate  $\gamma$  varies with an additional parameter: the Rossby number  $R_o$  of the vortex. The value of  $\gamma$  can vary non-monotonically with  $R_o$ , but always converges to QG theory as  $R_o$  approaches zero.

<sup>1</sup>D.A. Schechter, M.T. Montgomery and P.D. Reasor, J. Atmos. Sci., in press.

<sup>2</sup>L.J. Shapiro and M.T. Montgomery, J. Atmos. Sci. 50, 3322 (1993).

### A32B-0046 1330h POSTER

#### Conserving Mass in a CTM with a Pressure-Fixer: The Effect on a Full Ozone-Chemistry Run of IMPACT

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An important goal of multi-dimensional chemical tracer models (CTM's) is to illuminate the budgets of atmospheric species that are emitted, transported, and transformed. Necessary requirements for such models include:

1. Equality between the surface pressures inside the CTM and the surface pressures inside the source of its wind fields.
2. Maintenance of flat concentration fields for passive tracers.
3. Conserving tracer mass while advecting.

CTMs are often driven by meteorological fields (metfields) stored in a file (these are known as "off-line" CTM's). However, the winds in metfields are typically not guaranteed to be consistent with their accompanying surface pressure change because of effects from averaging, interpolation, and incompatible advection schemes. As a direct result of this inconsistency, it is impossible for the CTM to advect tracers using the given winds without sacrificing at least one of the three requirements above. Significant violations can occur, even though there may be no glaring effects in the modeled tracer distributions.

To address this problem, we have developed a "pressure-fixer" that adjusts the wind field to ensure the CTM surface pressure field follows the metfield surface pressure, and hence achieves all three of the requirements above. We have implemented such a pressure-fixer in IMPACT (our 3D combined stratosphere-troposphere CTM). We present here the effect of the pressure-fixer on the trace species simulated by a full ozone-chemistry run of both the troposphere and the stratosphere, including: O<sub>3</sub>, CO, and OH.

This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

### A32B-0047 1330h POSTER

#### A Comprehensive Validation of Global Chemistry Transport Models Against Observations for a Complete 4-year Simulation Period (1995 to 1998)

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As part of the EU funded project TRADEOFF (Air-craft Emissions: Contributions of various climate compounds to changes in composition and radiative forcing - tradeoff to reduce atmospheric impact) several European chemistry transport (CTM) and general circulation models (GCM) have been evaluated against observed tracer concentrations. The study focuses on the upper troposphere/lower stratosphere (UT/LS) region aiming to better assess the capability of the models to estimate the impact of aircraft emissions upon the atmosphere. For this purpose a comprehensive database of insitu observations of several compounds related to O<sub>3</sub> photochemistry was established for the 4-year period 1995 to 1998, covering observations from the most relevant commercial and scientific aircraft measurement campaigns and O<sub>3</sub> soundings. Tables of the positions and times of all observations available in a given month were supplied to the modellers and used to interpolate at every model time step the simulated tracer fields to the observation points. In this way a direct comparison of the models with observations taking into account the specific meteorological conditions during the measurements has been achieved. The models were run with a common database of surface and aircraft emissions (except for hydrocarbons) such that any differences between the models can be ascribed only to the different formulations of transport and chemistry. The results were evaluated with respect to the capability of the models to accurately simulate the observed concentration levels and variability, and the seasonal, latitudinal, and vertical distributions. The main results of this study will be presented.

URL: <http://www.lapeth.ethz.ch/~dominik/tradeoff/>

### A32B-0048 1330h POSTER

#### Isotopic enrichment of atmospheric carbonyl sulfide: constraining the contribution of carbonyl sulfide to the stratospheric aerosol layer

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We have assessed the contribution of carbonyl sulfide to stratospheric sulfate aerosol (SSA) by examining the OC34S and OC32S concentration profiles taken from infrared limb-transmittance spectra acquired with the JPL MkIV instrument, on balloon flights from various locations, during the period between 1991 and 2000. We found that OC34S is preferentially depleted with increasing altitude in a process that can be accounted for by solar photolysis. The derived 34S enrichment factor:  $e = +73.8 \pm 8.6$  permil, in conjunction with literature values of d34S approximately +11 permil for tropospheric OCS, and an approximately 10% net processing of OCS transported upwardly into the stratosphere, suggests that aerosol sulfate proceeding from OCS should be highly enriched in 34S. By comparing our prediction with previous reports of d34S of approximately +2.6 permil for background SSA, we infer that either OCS is a minor contributor to SSA or current views about its 34S-abundance and atmospheric circulation are seriously flawed.

#### A32B-0049 1330h POSTER

##### The University of Bern Ion Model: Modeling the Mixed $H^+(H_2O)_m(CH_3CN)_n$ Clusters in the Stratosphere

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Methyl cyanide ( $CH_3CN$ ) replaces water molecules in proton hydrates  $H^+(H_2O)_n$  at high rates in the stratosphere. Our global 2D ion model uses the latest stratospheric UARS MLS  $CH_3CN$  data together with neutral input from the NCAR SOCRATES model and computes mixed ion cluster  $H^+(H_2O)_n(CH_3CN)_m$  densities. The results are compared with in situ measurements from different sources.

#### A32B-0050 1330h POSTER

##### The University of Bern Ion Model: Time-Dependent Modeling of the Ions in the Stratosphere, Mesosphere and Lower Thermosphere

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The University of Bern Atmospheric Ion Model (UBAIM) is a time-dependent, pseudo-2D model of the ion chemistry in the earth atmosphere. It covers the latitudes between 85°S and 85°N and the (log-p) altitudes between 20 and 120km. On this grid, the system of differential equations describing the ion chemistry is integrated numerically until a dynamic equilibrium, governed by the diurnal changes in the incident radiation, is reached. Densities of main and minor atmospheric constituents are taken from the NCAR two-dimensional neutral SOCRATES model, the solar flux data are computed by the SOLAR2000 model. With this combination of models, it is possible to investigate the diurnal changes of the ion densities in the stratosphere, mesosphere, and lower thermosphere.

#### A32B-0051 1330h POSTER

##### Wave and Radiative Driving of the Lower Stratosphere: Trends

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Radiosonde temperature measurements at 100 hPa show cooling of both the tropical tropopause and the Arctic lower stratosphere in northern hemisphere winter. This is not consistent with a change in strength of the global-scale Brewer-Dobson circulation. It can be shown that Arctic temperatures (and the global-scale Brewer-Dobson circulation) are driven by dissipating planetary-scale waves which break in the extratropical stratosphere, while the tropical tropopause is driven by dissipating synoptic-scale waves which break on equatorial side of the subtropical jet. Analysis of vertical Eliassen and Palm (EP) flux through the extratropical tropopause shows that there has been a decrease in the contribution from planetary-scale waves, but an increase in the contribution from synoptic-scale waves. There has correspondingly been a decrease in EP-flux divergence in the extratropical stratosphere, but an increase in EP-flux divergence in the subtropics near the tropical tropopause. These changes in wave dissipation explain the cooling trends at the tropical tropopause and in the Arctic.

#### A32B-0052 1330h POSTER

##### Local Wave Driving of the Annual Cycle in Tropical Tropopause Temperatures

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Diagnostics from ECMWF analyses indicate that a significant factor in controlling the mean state and annual cycle of the tropical tropopause is breaking by synoptic-scale waves on the equatorial side of the subtropical jets. This wave breaking extends further into the tropics during northern hemisphere winter than northern hemisphere summer where it is displaced northward by the Asian monsoon. This annual cycle in wave breaking local to the tropopause explains the large annual cycle in temperatures just above the tropical tropopause and non-local wave breaking above 50 hPa is not important.

#### A32B-0053 1330h POSTER

##### Observation of Atmospheric Trace Species by Fourier Transform Infrared Spectrometer at Poker Flat, Alaska

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Communications Research Laboratory's (CRL) group has been promoting the Alaska Project, whose purpose is to investigate the Arctic middle atmosphere at Poker Flat, Alaska (65° N, 147° W). Eight kinds of observation instruments were installed. A Fourier transform infrared spectrometer (FTIR) was installed to observe minor constituents in the troposphere and stratosphere. This FTIR (Bruker 120HR) measures the solar infrared absorption spectrum with a spectral resolution of 0.0019  $cm^{-1}$  and a spectral range of 750 - 4200  $cm^{-1}$ . The absorption spectrum is used to derive total column amount and vertical profile of minor constituents.

Recently, the  $O_3$  annual trend was preliminarily obtained by retrieving  $O_3$  spectra from July 1999 to August 2001. The SFIT2 program developed by NIWA and NASA Langley was used for the retrieval. Each vertical profile of  $O_3$  was obtained from the absorption spectrum with a center frequency of 3044.6754  $cm^{-1}$  and a frequency resolution of 0.0035  $cm^{-1}$ . A spectral width of 0.1  $cm^{-1}$  was used to analyze the spectra. The absorption spectra are observed from February to November in the daytime. Selecting spectra with good signal to noise ratios, the spectra of 21 days were analyzed during 191 days (April 14 to October 21, 2000),

and the spectra of 64 days were analyzed during 188 days (February 24 to August 30, 2001). In the analysis of the spectra throughout two years, profiles of a priori, temperature, and pressure are fixed at the values of the standard atmosphere. The decreasing trend of  $O_3$  in the polar region from spring to summer is reproduced qualitatively. Analysis using more realistic profiles of temperature and pressure is in progress. Preliminary results for vertical profiles of HCl and related species will also be presented.

#### A32B-0054 1330h POSTER

##### Fractionation of $N_2O$ Isotopomers in the Simulated Stratospheric Sink Reactions

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Nitrous oxide ( $N_2O$ ) is an important trace gas in the atmosphere since it is radiatively active in the troposphere and also a precursor of nitric oxide which catalytically destroys ozone in the stratosphere. Natural isotope abundance in  $N_2O$  has been studied to understand its complex geochemical cycle. We previously reported that composition of  $N_2O$  isotopomers ( $^{14}N^{14}N^{16}O$ ,  $^{15}N^{14}N^{16}O$ ,  $^{14}N^{15}N^{16}O$ ,  $^{14}N^{14}N^{18}O$ ) has a unique vertical profile in the stratosphere such that heavier isotopomers are enriched and the intramolecular site preference of  $^{15}N$  for the center position increases with altitude. This enrichment was generally expected from the simulation experiments of photolysis at wavelengths of about 200 nm and theoretical calculations based on photo-induced isotopic fractionation effects. However, the detailed analyses of the vertical profiles showed several differences between observation and photolytic simulation/calculation.

Here we simulated another sink reaction, photo-oxidation of  $N_2O$  with excited oxygen atom,  $O(^1D)$ , to evaluate the fractionation of isotopomers in the process. Mixture of  $N_2O$  and  $O_3$  was irradiated by the light of 254 nm to initiate the photo-oxidation reaction, and isotopomer ratios of remaining  $N_2O$  were determined on the modified isotope-ratio-monitoring mass spectrometer that can analyze masses of molecular ion ( $N_2O^+$ ) and fragment ion ( $NO^+$ ). Factors controlling the stratospheric distribution of  $N_2O$  isotopomers will be discussed from fractionation factors during the sink reactions and stratospheric observations.

#### A32B-0055 1330h POSTER

##### Measurements of $O_2$ , $O_3$ and $NO_2$ During the MANTRA Balloon Campaign: Field-Testing the MAESTRO Space Instrument Concept

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The MAESTRO (Measurements of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation) satellite instrument will accompany the Atmospheric Chemistry Experiment Fourier Transform Spectrometer on the Canadian satellite SciSat-1 when

it is launched in December 2002. MAESTRO is a photodiode array spectrometer that will make solar occultation measurements of atmospheric attenuation during satellite sunrise and sunset to investigate the dynamical and chemical processes affecting stratospheric ozone. The precursor instrument to MAESTRO, the SunPhotoSpectrometer, was launched August 29, 2000 from Vanscoy, Saskatchewan as part of the main high-altitude balloon payload during the MANTRA (Middle Atmosphere Nitrogen TRend Assessment) 2000 field campaign. Sunrise occultation measurements made from a float altitude of 37 km have resulted in the retrieval of vertical profiles of ozone and nitrogen dioxide. Of particular relevance to the MAESTRO project are measurements of the A and B bands of molecular oxygen, which will be used for retrieving vertical profiles of atmospheric temperature and pressure on orbit. Molecular oxygen measurements made during the MANTRA campaign will be used to validate line-by-line models for MAESTRO temperature and pressure retrievals.

### A32B-0056 1330h POSTER

#### Attitude Determination for Limb-scanning Satellites: The "KNEE" at 305 nm

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<sup>3</sup>Odin web page, <http://www.snsb.se/Odin/Odin.html>

A technique based on observation and modelling of ultraviolet radiative transfer is developed to determine pointing to < 1 km accuracy for satellite instruments viewing Earth's limb. The method consists of locating the knee, defined as the maximum in the limb radiance profile, at a wavelength of 305 nm. The tangent altitude of this observational point (pixel or image, depending on whether or not the instrument is an imager) is known from radiative transfer modelling and is insensitive to tropospheric clouds, stratospheric aerosol, temperature, surface albedo and change in azimuth angle. Observations over several orbits with OSIRIS (optical spectrograph and infrared imager system) onboard the ODIN satellite and model calculations show the knee at 305 nm is at ~44 km. The insensitivity of the knee to these spatially variable geophysical parameters implies that the instrument measuring the limb radiance profile need not be an imager for this technique to be applicable. Although there is some sensitivity to SZA, ozone number density and pressure, it is predictable using appropriate model atmospheres and scattering geometry. Previously [e.g. McPeters et al., 2000], the knee at 345-355 nm was suggested for attitude sensing but our calculations and OSIRIS observations have shown this to be so sensitive to clouds and SZA that there is no knee in this wavelength range for certain scenarios. Internal scattering limits this technique with OSIRIS to wavelengths >300 nm. Odin is a Swedish-led satellite project funded jointly by Sweden (SNSB), Canada (CSA), Finland (Tekes) and France (CNES).

### A32B-0057 1330h POSTER

#### Stratospheric & Tropospheric Production of Nitrous Oxide: New Insights

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Nitrous oxide (N<sub>2</sub>O) is very important as a powerful greenhouse gas and the dominant source of stratospheric NO. Current uncertainties in its sources and sinks and the origin of its mass-independent isotopic enrichment warrant a discussion of a recent significant development.

• Since no production was seen by Estupinan et al (ESNW)<sup>1</sup> in photolysis of O<sub>3</sub>/O<sub>2</sub>/N<sub>2</sub> mixture at 532 nm, the excited O<sub>3</sub> capable of forming N<sub>2</sub>O (O<sub>3</sub>N<sup>2</sup>O) in Zipf-Prasad (ZP)<sup>2</sup> experiment is not produced in O(<sup>3</sup>P), O<sub>2</sub> recombination. It is likely a minor byproduct (reaction (1a)) of the termolecular quenching of O(<sup>1</sup>D) by O<sub>2</sub>.



O(<sup>1</sup>D) + O<sub>2</sub> + O<sub>2</sub> → O<sub>3</sub> (highly dissociative singlets) + O<sub>2</sub>\*# (1b)

The rate coefficient, k<sub>1a</sub>, needed to model ZP's quantum yield (Φ) of N<sub>2</sub>O is ~ 4x10<sup>-33</sup> cm<sup>-6</sup> s<sup>-1</sup>. The reaction (1b) or its parallel O(<sup>1</sup>D) + N<sub>2</sub> + O<sub>2</sub> → O<sub>3</sub> (highly dissociated triplets) + N<sub>2</sub> (R2) probably made a minor contribution to ZP's Φ since the lifetimes of the relevant excited O<sub>3</sub> is < 1 ns. Reactions (1b) & (2) amount to termolecular quenching of O(<sup>1</sup>D) since excited singlet and triplet O<sub>3</sub> dissociate.

• The Φ of N<sub>2</sub>O in ESNW and Maric & Burrows (MB)<sup>3</sup> experiments with 266/254 nm via the "true" N<sub>2</sub>O, O(<sup>1</sup>D) association may have been even lower than 3x10<sup>-7</sup>. Due to the reactivity of highly stretched molecules, the third generation N<sub>2</sub>O (N<sub>2</sub>O\*\*<sup>(3)</sup>) in the chain of events that produces N<sub>2</sub>O from the initial N<sub>2</sub>, O(<sup>1</sup>D) association may be prone to loss by O<sub>2</sub> (via possible reaction N<sub>2</sub>O\*\* + O<sub>2</sub> → N<sub>2</sub> + O<sub>3</sub><sup>\*</sup>) when n(O<sub>2</sub>) > n(N<sub>2</sub>). Thus, the idea that almost all of N<sub>2</sub>O observed by ESNW & MB was due to excited O<sub>3</sub> needs consideration. Their data suggest a smaller yield.

• Possibly, valuable information about the origin and properties of excited O<sub>3</sub> may be hidden under this difference. As the first step towards progress, we need to check if the differences in ZP's and ESNW's results can be narrowed by better experiments. ZP had used a spectrally coarse light source. ESNW got a larger yield of N<sub>2</sub>O than MB by reducing O<sub>3</sub>, and the residence time of the product N<sub>2</sub>O in the region affected by the photolysis. A rapid loss of N<sub>2</sub>O was observed by Black et al<sup>4</sup> in a set up where some products of the O<sub>3</sub> photolysis at 254 nm could react with N<sub>2</sub>O on the surface of reaction-chamber. Surface loss of N<sub>2</sub>O was probably not a concern in experiments at pressures > 20 atm and steel vessels. It could be a problem at 1 atm in narrow cylindrical vessels of pyrex with surface-to-volume ratio (S/V) > 1.

• More experimental studies of N<sub>2</sub>O production from O<sub>3</sub>N<sup>2</sup>O in air are therefore needed, using flowing gas (for lesser residence time) and reaction vessels with small S/V (for reduced surface loss). If O<sub>3</sub>N<sup>2</sup>O produces N<sub>2</sub>O then low-lying excited singlet and triplet states of O<sub>3</sub> may do the same in the denser troposphere (Prasad<sup>5</sup>). This possibility needs study since it may potentially compensate for the lesser efficiency of O<sub>3</sub>N<sup>2</sup>O in atmospheric production of N<sub>2</sub>O in the new scheme of O<sub>3</sub>N<sup>2</sup>O formation by only O(<sup>1</sup>D). Scanning of the entire O<sub>3</sub> absorption region with spectrally finely resolved irradiation is therefore also needed.

<sup>1</sup>Ph.D. Thesis, Georgia Tech, 2001; <sup>2</sup>GRL., 25, 4333, 1998; <sup>3</sup>J. Photochem. Photobiol. A, 66, 291, 1992; <sup>4</sup>J. Photochem., 22, 369, 1983; <sup>5</sup>JGR., 102, 21,527, 1997.

### A32B-0058 1330h POSTER

#### Observations of a Heterogeneous Source of OCIO from a Reaction of ClO with an Ice Surface

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Experiments presented in this contribution demonstrate a heterogeneous source of several chlorine oxides, including Cl<sub>2</sub>O, Cl<sub>2</sub>O<sub>3</sub>, and OCIO, from a flow of ClO radicals passed over glass or ice surfaces. ClO radicals were passed over these surfaces in a flow system monitored by UV-VIS absorption spectroscopy and Time-of-Flight mass spectrometry. It is suggested that a ClOH<sub>2</sub>O complex formed in the flow system is responsible for deposition of the ClO radicals onto the glass/ice surface, and several reaction pathways resulting in the formation of observed products are presented. The observation of OCIO evolving from an ice surface after ClO radicals have been introduced to it carries some possible atmospheric implications, as there is currently a missing source of OCIO in the chemically perturbed polar vortex. The ClO/BrO reaction system is currently believed to be the only source of OCIO in the stratosphere, and several studies show this reaction system to severely underestimate OCIO production in the polar vortex. It is suggested this new heterogeneous source of OCIO from a ClOH<sub>2</sub>O reaction with an ice surface could carry implications on the total OCIO budget.

### A32B-0059 1330h POSTER

#### Raman Spectroscopy and Microphysics of Single PSC Precursor Particles Suspended in a Quadrupole Trap

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Polar stratospheric clouds (PSCs) consist primarily of solid nitric acid trihydrate (NAT) particles, which are thought to nucleate via HNO<sub>3</sub> uptake on background sulfuric acid particles at temperatures below 195 K. The mechanism for this process is uncertain, and depends on whether the sulfuric acid particles are solid or liquid at these temperatures. Previous results from laboratory and field measurements are mixed; our previous single-particle laboratory experiments showed that binary H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O particles at stratospheric compositions are essentially metastable in the liquid phase when cooled to PSC temperatures. Currently, we are investigating the detailed microphysics of binary (H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O) and ternary (HNO<sub>3</sub>/H<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>O) single particles suspended in an electrodynamic levitator, using optical elastic scattering and Raman spectroscopy to observe changes in phase and composition. Single-particle Raman spectra for supercooled binary particles exhibit spectral distributions which alter markedly with decreasing temperature down to 190 K. The variations signify increasing dissociation of HSO<sub>4</sub>(-) to SO<sub>4</sub>(-) with decreasing temperature, consistent with measurements for bulk solutions. Upon gradual warming of supercooled liquid binary particles, some of them freeze briefly in a narrow "window" of the phase diagram, near 210 K and 60 weight per cent H<sub>2</sub>SO<sub>4</sub>. We will discuss the Raman spectroscopy and microphysical behavior of the liquid and frozen particles for both the binary and ternary systems.

This research was supported by the NASA Atmospheric Effects of Aviation Program.

### A32C MC: 123 Wednesday 1330h

#### Predictability of the North American Monsoon System I

Presiding: R Oglesby,  
NASA/MSFC/GHCC

### A32C-01 1330h INVITED

#### Modelling and Predicting the North American Monsoon System: Status and Prospects

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The regional summer climate regime across South-west North America provides an extreme challenge to numerical models. Precipitation in this semiarid monsoonal domain is modulated by a daunting combination of complex topography, ocean-atmosphere and land-atmosphere fluxes, midlatitude synoptic systems, organized mesoscale convective processes, and subgrid-scale deep convective clouds. To add to this complexity, verification data needed to facilitate model simulation improvement are sparse across much of the domain. This talk discusses some recent North American Monsoon System (NAMS) simulations and outlines a strategy for progress on modeling and predicting the NAMS, to take place in concert with field activities associated with the upcoming North American Monsoon Experiment (NAME).

### A32C-02 1350h

#### Simulating Extreme Summer Precipitation Patterns in the North American Monsoon Region using the CCM3/HRBATS Model

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Current climate integrations with the National Center for Atmospheric Research Community Climate Model (CCM3) show a very pronounced dry bias in summer precipitation over the North American Monsoon System (NAMS) region. Additionally, summer