

A51G-02 1040h INVITED

First Stage of Upper-stratospheric Ozone Recovery

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Global ozone trends derived from the Stratospheric Aerosol and Gas Experiment I and II (SAGE I/II) combined with the more recent Halogen Occultation Experiment (HALOE) observations provide evidence of a slowdown in stratospheric ozone losses since 1997. This evidence is quantified by the cumulative sum of residual differences from the predicted linear trend. The cumulative residuals indicate that the rate of ozone loss at 35-45 km altitudes globally has diminished. These changes in loss rates are consistent with the slowdown of total stratospheric chlorine increases characterized by HALOE HCl measurements. These changes in the ozone loss rates in the upper stratosphere are significant and constitute the first stage of a recovery of the ozone layer.

URL: http://vortex.nsstc.uah.edu/atmchem/recent_events/upperstrat03_recovery.html

A51G-03 1100h INVITED

Measurements of water and its isotopologues: insights into the atmospheric hydrological cycle

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HALOE, POAM, ATMOS, and SAGE have all reported measurements of H₂O (and in several cases, H₂O isotopologues) from orbit by solar occultation. I will discuss recent measurements pertaining to trends in stratospheric and upper tropospheric H₂O as well as describe the use of isotopic analysis to constrain the mechanisms that control the humidity of this region of the atmosphere.

A51G-04 1120h

Data Assimilation of SAGE II Data: Solar and Volcanic Effects

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In an earlier published paper, we showed the data assimilation results for ozone using the SAGE II solar occultation data and a two-dimensional chemistry-transport model. In this earlier work, no solar cycle UV dependence was included in the model. Here, we show results using this same model, but now including solar cycle UV variations. These new results are in better agreement with the independent TOMS data, particularly during the middle and late 1980s. The new results also show lesser episodic ozone decreases in the lower stratosphere that are associated with the El Chichon (3% compared to 4%) and Mt. Pinatubo (6% compared to 9%) eruptions enhancements in stratospheric aerosol. Comparisons are shown between ozone profiles

from the data assimilation and from the SAGE II data to illustrate both the "goodness" of fit of the assimilation ozone profiles to the input data and also to clearly see the differences in the signature of the volcanic and solar effects on the ozone profiles.

A51G-05 1135h

The Impact of the 1991 Pinatubo Volcanic Eruption on Climate Using a Vertically Resolved Stratospheric Aerosol Data Set Derived from SAGE II Observations

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Satellite observations of stratospheric aerosol extinction with the SAGE I, SAGE II, and SAM II instruments provide the best global coverage for the past two decades. The SAGE III instrument will continue this outstanding monitoring program. In addition SAGE provides the vertical structure of aerosol characteristics, which is important for calculation of aerosol radiative forcing and radiative heating of the lower stratosphere. Therefore these observations are invaluable for simulating the climate effects of stratospheric aerosols and their contribution to contemporary climate change. Most of our understanding of the impacts of volcanic eruptions comes from studies of the 1991 Mt. Pinatubo eruption in the Philippines, the largest and best observed eruption of the 20th century. Using SAGE II and CLAES/ISAMS satellite observations we have developed a spectral-, space-, and time-dependent set of aerosol parameters for two years after the Mt. Pinatubo eruption. Here we use this aerosol data set to study the effect of this eruption on the Arctic Oscillation (AO), accounting for the quasi-biennial oscillation (QBO) in the tropical stratosphere for the first time. We use the SKYHI troposphere-stratosphere-mesosphere climate model, which effectively assimilates observed zonal mean winds in the tropical stratosphere. The dynamical response to the Pinatubo eruption, with an enhanced positive mode of the AO, was reproduced by the model in the first and second Northern Hemisphere winters following the eruption, as observed. The phase of the QBO modulates climate system sensitivity to an external forcing. The QBO in its westerly phase strengthens the AO response. Because of nonlinear interactions, aerosols and the QBO together produce a stronger response than a linear superposition of responses to each of these forcings. Improved quantification of the QBO effect helps to better understand mechanisms of the stratospheric contribution to natural and externally forced climate variability.

A51G-06 1150h

Seasonal And QBO Variations Of Ascent Rate In The Tropical Lower Stratosphere As Inferred From UARS HALOE Trace Gas Data

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Seasonal and interannual variations in ascent rates are investigated as a function of latitude and height, using water vapor (H₂O) and methane (CH₄) data from the stratospheric measurements of the Halogen Occultation Experiment (HALOE). The ascent rate is inferred from the ascending signal of variations in the entry value of [H₂O] + 2[CH₄] (H). Within ±15° of the equator, the derived ascent rate exhibits mainly two kinds of dominant variations with a clear latitudinal structure, seasonal variation and the quasi-biennial oscillation (QBO). The seasonal cycle exhibits a vertically in-phase variation, with a northern winter maximum of 0.2-0.4 mm s⁻¹ and a summer minimum of ~0.2 mm s⁻¹ in the 20-60 hPa layer. The latitudinal structure is characterized by an early appearance of a subtropical summer maximum of the ascent rate and by double peaks at 10-15°N and S during the northern winter season. The QBO component of the ascent rate shows tropically confined anomalies with a rapid downward propagation, but mass attenuation anomalies estimated from the ascent rate show a much slower downward propagation. The descent anomalies exhibit a well-structured and equatorially symmetric variation, while the ascent anomalies have a tendency to propagate latitudinally. This might be connected with the phase dependency of the QBO acceleration. An examination of the phase and amplitude of the ascent rate and temperature for both the seasonal and QBO components emphasizes that the radiative damping timescale is considerably long (40~100 days) below 40 hPa.

A51G-07 1205h

Stratosphere-Troposphere Exchange studies using SAGE II data

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Stratosphere-Troposphere Exchange (STE) is of significant importance to the chemistry and dynamics of the troposphere and the stratosphere yet many aspects are not well understood. Stratospheric Aerosols and Gas Experiment II (SAGE II) version 6.2 data, with improved water vapor, near global coverage, and excellent vertical resolution, is ideal to investigate stratosphere-troposphere exchange. The SAGE II 6.2 water vapor and ozone data together with United Kingdom Meteorological Office (UKMO) assimilated data are utilized to identify air of tropospheric characteristics in the stratosphere and air with stratospheric characteristics in the troposphere. Air that may be of stratospheric origin residing in the troposphere is identified by high ozone and low water vapor mixing ratios relative to tropospheric averages while tropospheric air in the stratosphere is identified by high water vapor and low ozone content relative to the stratospheric averages. For these determinations, the cold point tropopause is utilized in the tropics and the 3.5 x 10⁻⁶ m² s⁻¹ K kg⁻¹ potential vorticity surface defines the tropopause in the mid-latitudes. The frequency of stratosphere-troposphere exchange is calculated globally and regionally to determine STE activity and the vertical structure of the exchange is investigated. In this paper, the data analysis approach, results, and interpretations of this investigation will be discussed.

A51H MCC: 3018 Friday 1020h

Use of Inverse Modeling for Constraining Global Budgets of Atmospheric Trace Gases II (*joint with B*)*Presiding:* A M Michalak, NOAA

Climate Modeling and Diagnostics Laboratory; W Peters, NOAA Climate Modeling and Diagnostics Laboratory; P P Tans, NOAA Climate Modeling and Diagnostics Laboratory

A51H-01 1020h INVITED

Estimation of Trace Gas Fluxes by Inverse Modelling

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A wide range of scientific questions regarding chemically and/or radiatively important trace gases necessitate determinations of their sources and sinks at local to global scales. A powerful method for such determinations involves solution of an inverse problem in which the observed concentrations are effectively Lagrangian line integrals and the unknown sources or sinks are contained in the integrands. The inverse problem consists of calculating optimal estimates of the unknowns in the Bayesian sense using an atmospheric transport model and trace gas measurements gathered over space and time. Great care is necessary to include the effects of both measurement and transport model errors in calculating the uncertainty in the optimal estimates. We review the results of recent studies which use three-dimensional Eulerian (specifically MATCH) or Lagrangian transport models and Kalman filter and other optimization methods to compute emissions of methane, nitrous oxide, and selected halocarbons. These studies use high frequency trace gas observations from global networks (AGAGE, CMDL) to calibrate a priori emission maps for particular processes and geographic regions. The methods allow estimation of time varying emissions. For the hydrogen-containing gases these emission estimates require accurate specification of the concentrations of the hydroxyl radical which constitute their major sink. Hydroxyl radical levels can be optimally estimated in a separate problem using measurements of methyl chloroform whose global emissions are already very well known. The results show that the inverse approach is a powerful complement to traditional surface flux aggregation methods. At the same time, the inverse approach has its own limitations associated especially with transport model errors and/or inadequate atmospheric measurements.

A51H-02 1035h

Estimating Global Nitrous Oxide Surface Fluxes Using a Time-Dependent Bayesian Inversion Technique

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The continual increase of nitrous oxide in the atmosphere, combined with its strong greenhouse warming and stratospheric ozone-depleting properties, make improved understanding of the atmospheric nitrous oxide budget an important scientific goal. During the period 1998 to 2002, nitrous oxide measurements from the NOAA/CMDL global flask network showed significant inter-annual variability in both the global atmospheric growth rate and the latitudinal gradient at the surface. Additionally, measurements from stations located in high northern and southern latitudes exhibited seasonal cycles with amplitudes exceeding 0.3 ppbv. To explore the information about the global nitrous oxide budget contained in these seasonal and inter-annual patterns, we implemented a time-dependent Bayesian inversion with the NOAA/CMDL flask measurements and the chemical transport model TM3. Monthly surface fluxes were estimated for the 22 basis regions defined by the TRANSCOM project, using a priori fluxes from the GEIA inventory which includes both natural and anthropogenic contributions. The presentation will cover both the numerical methodology and the inversion results.

A51H-03 1050h

INTERANNUAL METHANE SOURCES INFERRED BY INVERSION OF ATMOSPHERIC TRANSPORT

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A primary goal of developing the methane (CH₄) atmospheric measurement network is to better characterize the sources of the second anthropogenic greenhouse gas in space and time. Chemical transport models (CTM) can be used to analyse atmospheric measurements in terms of surface fluxes using inverse methodology. The main sink of methane, chemical removal by OH radicals, is also an unknown of the inverse problem that can hardly be optimized using surface CH₄ observations alone. We have developed a double 3D atmospheric inversion of both methyl Chloroform (CH₃CCl₃) and methane atmospheric observations in order to optimize interannual OH concentrations and CH₄ surface sources successively, for the 1984-1997 period. The inversion scheme infers CH₄ monthly sources and sinks from GLOBALVIEW-CH₄ observations and GAGE-AGAGE CH₃CCl₃ observations using INCA-LMDZ CTM in forward and backward (self-adjoint) modes. Natural tropical ecosystems are found to explain most of the interannual variability (IAV) of CH₄ sources over the period. In this presentation, We illustrate this result and present contribution to IAV of all types of emissions, at a continental scale. Several sensitivity inversions are also presented to quantify the influence inversion set up on inferred sources : OH optimization, stations used, observation error, flux aggregation error,

A51H-04 1105h

Global Carbon Fluxes Based on Global CO₂ and Carbon Isotope Measurements

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Obtaining detailed and accurate estimates of carbon fluxes is of considerable scientific and political importance, and using isotopic information can help constrain these fluxes. For example, measurements of the carbon isotope ratio of CO₂ ($\delta^{13}\text{C}$) helps resolve longitudinal structure in carbon fluxes by constraining the land-sea partitioning of regional carbon fluxes. In this study, we improve on past efforts to use $\delta^{13}\text{C}$ measurements to further constrain CO₂ fluxes in several important ways. First, we use more observations of $\delta^{13}\text{C}$ than have been used in the past. There are now 51 stations from which regular measurements of $\delta^{13}\text{C}$ are available. Secondly, we use a monthly resolution time-dependent inversion technique, which allows us to make maximal use of the atmospheric $\delta^{13}\text{C}$ data. Specifically, we take advantage of the information contained in the seasonal cycles of CO₂ and $\delta^{13}\text{C}$, not just the annual mean values. Although we use the best possible estimates of the terrestrial isotopic signatures and isotopic disequilibrium terms, there is significant uncertainty associated with these terms, and we will address the ways in which their uncertainty limits the ability of $\delta^{13}\text{C}$ to add useful information to the inversion. In this presentation, we will also introduce the technique that we use for atmospheric inversions, a fixed-lag Kalman smoother. This technique offers computational efficiency while also having the advantage of propagating covariance forward in time.

A51H-05 1120h

A Joint Atmosphere-Ocean Inversion for Surface Fluxes of Carbon Dioxide

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An inversion method has been developed for incorporating both atmospheric and oceanic constraints on surface fluxes of carbon dioxide. Previous estimates of the air-sea flux of carbon dioxide using only atmospheric constraints predict different ocean surface fluxes than do inversions of only ocean interior data, particularly in the temperate Pacific ocean. We simultaneously invert atmospheric CO₂ concentrations and estimates of anthropogenic and preindustrial carbon concentrations from the ocean interior to produce a consistent estimate of surface fluxes. Sensitivity to transport uncertainty is studied using atmospheric transport estimates from TransCom3 and a suite of ocean GCM simulations.

A51H-06 1135h

A 4-D Variational Data Assimilation Approach for Estimating Time-Varying Sources and Sinks of CO₂

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Inversions of the historical atmospheric CO₂ measurement record provide robust estimates of CO₂ sources and sinks at generally coarse scales: at continental and ocean basin scales in space, and at seasonal to monthly time scales. To help choose between the different possible physical mechanisms driving the sources and sinks, finer-scale results are desired ? e.g., daily-average fluxes at the resolution of 100s of km. Global inversions at these scales are currently computationally feasible only using adjoint methods. Using the adjoint of a global 3-D atmospheric transport model (with 2x2.5 degree resolution), we present daily CO₂ flux estimates at the model resolution using a variational data assimilation method (4-D Var). With two different sets of modeled surface CO₂ fluxes (LPJ land/NCAR ocean vs. CASA land/Takahashi ocean), we perform observing system simulation experiments (OSSEs), both in a perfect model context and using two different transport models, to assess the total errors expected at these fine scales when using CO₂ observations from a greatly expanded version of our current network (no satellite-based measurements, but with hourly-resolved measurements from hundreds of in situ analyzers). In the perfect model case, the OSSE total errors show corrections at coarser scales, but persistent errors at finer spatial scales. Apparently, the inversion can improve the fluxes only at scales dictated by the density of the observations, rather than the underlying resolution of the solution method. [This result implies, for example, that a network equivalent to 100 fixed sites across the continental U.S. would be limited to a resolution of about 400 km, at best.] The OSSE errors for the inversion using different transport models suggests that even this resolution limit is quite optimistic. A dense observing network, coupled to a transport model that accurately represents synoptic-scale (perhaps even meso-scale) mixing, is required; the correlations assumed in the inverse procedure must be tuned well, also. The adjoint transport model is used, independent of the inverse procedure, to show that the surface fluxes for the mid- to high-latitudes are constrained much better than the tropical fluxes by the assumed network. This suggests that we will have to be satisfied with much coarser resolution in the tropics, unless a highly dense observing network is established there.

A51H-07 1150h

A Quantitative Method to Incorporate Atmospheric Transport Errors in Inverse Modeling Studies

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Previous inverse studies have pointed to the sensitivity of flux estimates to errors in atmospheric transport for tracers such as CO₂. Current inverse approaches either neglect transport errors or only attempt to roughly assess their effects from the spread in results derived using different transport models or from "high-frequency variability" in observed tracer concentrations. We describe a method to quantitatively account for transport errors in inverse analyses by incorporating uncertainties in mean winds into the stochastic motions of air parcels. The magnitude of errors in wind fields, as well as their spatiotemporal covariances, are determined by direct comparison of assimilated winds to radiosonde observations. The transport error statistics established by the comparisons are propagated through the stochastic motions of air parcels as simulated by the Stochastic Time-Inverted Lagrangian Transport (STILT) model. We illustrate this method by using atmospheric CO₂ observations over the continent and examine the effect of transport errors on estimates of regional terrestrial carbon fluxes.

A51H-08 1205h

Sensitivity of inverted carbon sources and sinks from seasonally and interannual varying fossil fuel emission estimates

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The possibility of a ?fossil fuel rectifier? is tested using two models (PCTM and MATCH:NCEP) that span the spectrum of vertical trapping and rectification as established in the TransCom 3 Atmospheric Carbon Inversion Intercomparison Experiment. Tests with a pseudo fossil fuel emissions field with varying seasonal amplitude suggests that a fossil fuel rectifier is likely to be a small factor in annual mean inversions. However, seasonality in the fossil signal and/or interannual variations in the spatial distribution has a significant impact on seasonal and interannual inversions. Though the level of seasonality in this emissions field is not well known, a 30% amplitude modification case in this exercise closely matches a recently suggested fossil amplitude derived from observational data. Inverse results including this level of fossil seasonality result in posterior flux estimates differing by up to 50% in the northern temperate land regions where the majority of the hypothesized northern land sink is located. This will vary with the extent to which simulated transport vertically traps surface fluxes as evidenced by the results from the two models chosen for this sensitivity study. The impact of interannual variations in the surface pattern of fossil fuel emissions have also been explored. In this experiment, inverse results which attempt to include regional shifts in fossil fuel over the last twenty years will be compared to a case in which the surface emission patterns are fixed. Exclusion of seasonal and interannual fossil fuel variations in inversion studies may lead to misinterpretation of results. This holds particular importance given that the proposed northern land sink and large fossil fuel emissions are coincident in space. Better representation of these variations is essential to maintain accuracy in inverse results as atmospheric inversion studies move to greater spatial and temporal resolution.

A52A MCC: Level 2 Friday 1330h

Isotopic Constraints on Global Budgets of Atmospheric Gases III Posters (joint with B, H, OS, PP)

Presiding: K A Boering, University of California, Berkeley; N Jones, University of Wollongong

A52A-0761 1330h POSTER

Regional Impact of Terrestrial Ecosystem on Global Carbon Cycle: Indicated by Isotopic Measurements of Atmospheric CO₂ at Boreal Forest (Fraserdale, Canada)

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Eight intensive campaigns were conducted in different seasons over a 3-year period (1998 to 2000) at a boreal forest site, Fraserdale (50°N, 81°W) in northern Ontario, Canada. Flask air samples were taken both at the 20-m level of a 40-m tower and from aircraft profiles (six campaigns), and were analyzed for both CO₂ concentration and their isotopic compositions ($\delta^{13}C$ and $\delta^{18}O$). The results from these intensive campaigns are compared with Marine Boundary Layer (MBL) data at the same latitude, derived from the observations obtained by the CMDL/NOAA cooperative air sampling network, in order to understand the impact of regional sources and sinks on global carbon cycle. The amplitude of variation in each ground campaign and the difference between the campaigns and MBL show typical seasonal patterns for both CO₂ concentration and isotopic compositions, with smaller values in the winter, and larger values in the summer and Fall, reflecting the strength and balance of regional sources/sinks driven by ecosystem processes. The most interesting discovery is that the minimum value of $\delta^{18}O$ from Fraserdale campaign data occurs in the September - October period, which does not match the minimum value of $\delta^{18}O$ in local precipitation (the latter occurs in December - January) but matches the pattern of MBL $\delta^{18}O$ in mid-high latitudes of the Northern Hemisphere. This indicates Sept. and Oct. are a very active time of year for CO₂ isotope exchange with soil water and that $\delta^{18}O$ of the respiration flux from the local ecosystem has a significant imprint on $\delta^{18}O$ in global background air (MBL). Similar comparisons are also applied to aircraft flask samples. Less variation (including daily and seasonal changes) and less difference between campaigns and MBL are found in aircraft data; and MBL and the aircraft data closely agree with each other. Comparison of ground and aircraft campaign data with MBL compositions reveals very valuable information for understanding the processes of propagation of terrestrial biosphere signals into global background values of the atmospheric compositions (CO₂, $\delta^{13}C$ and $\delta^{18}O$), and the impact of regional sources and sinks on global carbon cycle.

A52A-0762 1330h POSTER

Sensitivity of terrestrial ecosystem models of carbon isotope ratios to changes in vegetation properties and meteorology

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We look at the sensitivity of simulated carbon isotope ratios and CO₂ fluxes from several terrestrial ecosystem models to variations in vegetation properties and meteorology. In particular, we compare results from the Community Land Model (CLM2.1); and two versions of the Simple Model of the Biosphere (SiB2.5 and SiB3), to changes in vegetative land cover data sets: Fourier-Adjustment, Solar zenith angle corrected, Interpolated Reconstructed Normalized Difference Vegetation Index (FASIR-NDVI); Global Inventory Monitoring and Modeling Studies NDVI (GIMMS-NDVI); and the CLM Plant Functional Type (PFT) classification scheme, and meteorology: European Center for Medium Range Forecast (ECMWF); National Center for Environmental Prediction (NCEP); and Community Climate Model (NCAR-CCM3). We compare spatial distributions of d13C of terrestrial plant carbon, the zonal gradient of d13C values, and time-series of mean annual d13C values. We also examine the response of the models to changes in meteorology (relative humidity and precipitation) and recycling of canopy CO₂. Finally, we compare simulated seasonal cycles of d13C of atmospheric CO₂ and the north-south d13C gradient to observed values at several NOAA Flask Stations using response functions (transport fields) developed in Transcom. Results highlight the importance of specified precipitation and precipitation anomalies to carbon isotope discrimination resulting from physiological stress.

A52A-0763 1330h POSTER

Disturbance-Induced Changes in the Oxygen Isotopic Composition of Atmospheric CO₂ at High Northern Latitudes

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At high northern latitudes, interannual variation in the seasonal cycle of atmospheric C¹⁸O exceeds that observed for ¹³CO₂ or CO₂. Shifts in species composition or climate may contribute to this variability in C¹⁸O by influencing the timing of photosynthesis and respiration and the cycling of meteoric water. We hypothesized that increased forest fire frequency contributes to variability in C¹⁸O by changing surface energy partitioning, fluxes of water and CO₂, and vegetation structure and function. We measured the $\delta^{18}O$ of ecosystem water pools (leaf, stem, soil, water vapor, and precipitation) in an 80 year old Picea mariana stand and a 15 year old Populus tremuloides stand in the Alaskan interior. The two forests displayed similar $\delta^{18}O$ values for precipitation, soil, and stem water, however mid-day leaf water in P. mariana was consistently more enriched in ¹⁸O than that of P. tremuloides on the order of 2 ‰. We predicted differences in diurnal and seasonal C¹⁸O fluxes between sites by incorporating net CO₂ eddy flux and micrometeorological measurements with oxygen isotopic signatures of ecosystem water pools, local background atmospheric CO₂, and nighttime ecosystem respiration. We then used a simple age-distribution model of boreal forests to predict changes in the shape of the seasonal cycle of C¹⁸O that would accompany shifts in the disturbance regime.