August 14, 1998

Proposed Augmentation for AEAP Participation in SOLVE

The next major field mission under the Upper Atmosphere Research Program (UARP) will be the SAGE III Ozone Loss and Validation Experiment (SOLVE), which is scheduled for northern hemisphere Winter, 1999-2000. SOLVE is a proposed Arctic DC-8, ER-2 aircraft and balloon measurement campaign designed to examine the processes which control polar to mid-latitude stratospheric ozone levels. The mission will also acquire correlative measurements needed to validate the Stratospheric Aerosol and Gas Experiment (SAGE) III satellite mission and will use these satellite measurements to help quantitatively assess high latitude ozone loss. The SOLVE mission planning document is attached (or may be obtained through http://www.hq.nasa.gov/office/ese/nra/nra98oes08/index.html).

SOLVE is co-sponsored by UARP, the Atmospheric Chemistry Modeling and Analysis Program (ACMAP), and Earth Observing System (EOS) of NASA's Earth Systems Enterprise as part of the validation program for the Stratospheric Aerosol and Gas Experiment (SAGE) III instrument. Because several of the central scientific objectives of SOLVE are intimately linked to key questions in assessment of High Speed Civil Transport (HSCT) impacts, the Atmospheric Effects of Aviation Project (AEAP) is expected to participate in SOLVE. Participation in SOLVE will particularly help AEAP answer pressing questions about polar processes, reactive nitrogen partitioning, and transport in the winter high latitude regions. Under the current planned budget for FY00, AEAP will be able to support SOLVE at only a moderate level, approximately $500k through the Atmospheric Effects of Stratospheric Aircraft (AESA) project within the High Speed Research Program (HSRP). This contribution will primarily fund theory teams and data analysis which will examine SOLVE data for its implications regarding HSCT assessments. Here we propose to augment AEAP funding by $3M to become a full partner in SOLVE with UARP in pursuit of aviation-specific science objectives.

Science Motivation

Overall, the SOLVE mission is motivated by the need to understand the apparent decreasing trend in Northern hemisphere polar ozone, which has culminated in record low ozone amounts in recent years [Newman et al., 1997]. The dynamical and chemical causes must be better characterized in order to predict whether the recent record low ozone is part of a long-term trend or a short term

variation. We need to know how ozone will respond to decreasing chlorine loading resulting from international protocols on chlorofluorocarbons, while at the same time stratospheric dynamics are altered by global climate change from increasing greenhouse gases. The current SOLVE science plan is designed to address these issues.

The science objectives of SOLVE are in many cases complimentary to, or the same as, those needed to improve our assessment of the impact of a fleet of HSCT aircraft on stratospheric ozone. We need to understand the current polar stratosphere better in order to better assess the potential effects of HSCT emissions, which may be substantial in this region (more below). In fact, the SOLVE plan was designed with HSCT assessment issues in mind. The SOLVE plan is, however, very ambitious and not all of the possible implementation options will be possible for financial and practical reasons. In order to increase the science yield, SOLVE management and its science leadership are willing to expand the core program to address HSCT issues more forcefully. This presents AEAP the opportunity to obtain critical research data and analysis for a fraction of the total cost for a major field expedition. We propose to augment the SOLVE mission deployment to pursue data focused on specific aircraft assessment-driven issues, which together with the entire SOLVE data set will significantly enhance confidence in HSCT assessment calculations. This work will address the issues of HSCT exhaust transport in polar regions; NOy partitioning and O3 loss kinetics; the impact of HSCT-emitted NOx, H2O, and particles on polar processes; and the possible contribution of aircraft-produced particles to the lower stratospheric aerosol burden. AESA science motivation and potential SOLVE contributions follow.

Transport

Uncertainty in understanding transport processes in the stratosphere and their implementation in numerical assessment models has been identified as a major contributor to the overall uncertainty in predicting HSCT impacts [Stolarski, et al., 1995; Kawa et al., 1998]. Transport processes control the global dispersion of HSCT emissions, their residence time, and, hence, their concentration in various regions of the atmosphere, which in turn produces the chemical effect on ozone. Incomplete theoretical understanding, limitations of computer parameterizations, and lack of diagnostics for comparison with reality make a fully quantitative evaluation of this uncertainty impossible. Implementation of transport processes has, however, been identified as a considerable source of the difference among models in their predictions of the HSCT impact, which is one component of the total uncertainty.

Field observation campaigns have provided measurements for first-order diagnosis of transport circulations as well as for model evaluation. The Stratospheric Tracers of Atmospheric Transport (STRAT) mission (jointly with UARP and ACMAP) measured the morphology of long-lived tracers and dynamical quantities as functions of altitude, latitude, and season in order to help determine rates for global-scale transport and to provide high quality observations for testing assessment models. STRAT completed an extensive set of measurements from the NASA ER-2 aircraft at tropical and mid latitudes. Analysis of these data has determined the mean age of the air across the northern hemisphere lower stratosphere and its seasonal dependence. This is an important diagnostic for understanding the lifetime of HSCT exhaust. In addition, STRAT data has permitted calculation of the rate of transport of air from mid-latitudes, where most HSCTs would fly, into tropical latitudes, where emissions could become entrained into the climatological upward circulation and transported to higher altitudes where chemical sensitivity is greater.

In conjunction with STRAT, the Observations from the Middle Stratosphere (OMS) measurements from high altitude balloons were carried out to complement measurements by the ER-2 and satellites. Such middle stratosphere observations fill the critical measurement gap in altitude from the ER-2 at 20 km up to approximately 30 km. These measurements extend the age of air calculations to higher altitudes and allow estimates of transport rates as a function of altitude. Such high altitude measurements are essential to evaluating global transport models. The combination of STRAT and OMS data for age of air is an entirely new and powerful diagnostic test for evaluating model simulations of atmospheric transport. Quantitative estimates for mid-latitude and tropical transport from tropospheric source gases and CO_2 also provide constraints on model formulations which have not been available before.

Using STRAT and OMS data, the Models and Measurements II (M&M II) analysis has shown that uncertainty in transport processes and representation in models continues to be a major problem in HSCT assessment. Comparisons with the new measurement diagnostics have exposed a number of model shortcomings, most notably age of air simulations (Figure 1). In addition, model intercomparison within the Global Modeling Initiative (GMI) and M&M II has shown a wide range of variability for predictions of HSCT exhaust accumulation and dispersion in the stratosphere (Figure 2). The causes of these discrepancies, and hence corrective measures, however, are not yet apparent. Age of air is correlated to HSCT exhaust dispersion among models but the mechanistic basis for this relationship is not clear, nor is it known quantitatively how either of these diagnostics is linked to column ozone perturbations. Transport controls the background NOy, Cly, and other tracers against which the HSCT perturbation is superimposed. This background determines the magnitude of the HSCT perturbation to a large extent and thus is very

important to model correctly. The sensitivity is especially acute in the polar regions where highly non-linear reactions control partitioning and ozone loss rates.

The proposed AEAP augmentation to SOLVE will include targeted deployment of balloon tracer instrumentation (e.g., OMS) to provide age of air and other diagnostics at high latitudes in late fall, winter, and possibly early spring. These measurements will enable analysis of the descent circulation which will act to both supply HSCT exhaust into the region via the large scale diabatic circulation as well as flush exhaust from the region which accumulates during summer or is emitted directly into the vortex in the winter. The balloon measurements, in concert with the horizontal transects from aircraft measurements, will enable estimates to be made of the inward mixing of mid latitude air (where most HSCTs will fly) to the vortex. AEAP support of SOLVE will also ensure that the ER-2 payload includes a full complement of needed trace gas measurements (which would not be included in the minimum SOLVE payload). Based on previous mission experience the cost of the additional balloon and aircraft tracer measurements is approximately $1M. Such quantitative estimates for the strength of the supply and removal components of the exhaust transport circulation are necessary to accurately assess the impact of the HSCT emissions on ozone chemistry in polar regions.

Chemical Reaction Kinetics

Uncertainty in HSCT impacts resulting from errors in modeling of atmospheric chemical processes is propagated through the dependence of ozone and other chemical changes on the change in nitrogen oxide gases and water vapor produced by the HSCT emission. Chemical processes involving particles are discussed below. Uncertainty in the total ozone perturbation from HSCTs due to chemical kinetics was evaluated to be +/- 1% [Solarski et al., 1995]. Additional chemical uncertainties are attributed to differing reaction schemes and implementation in different models. Because of the small abundances of critical species and difficulty of measurements, the possibility of unknown chemical processes cannot be dismissed either.

Although the kinetics uncertainty of +/- 1% is substantial, our overall assessment of chemical processes is relatively good [Kawa et al., 1998]. The kinetics error is quantifiable (unlike transport for example). In some conditions, atmospheric measurements indicate that the laboratory-based error estimates are even overly conservative [Cohen et al., 1994]. The M&M II comparison of model chemical simulations has produced a very positive result. Chemical integration of the various models, constrained by data, yield very similar results. This implies that different chemical responses among models may be attributed largely to differences in transport

and possibly multi-phase processes. Chemical integration is not a large source of model variance. In addition, comparisons with measurements of reactive species, mostly at mid latitudes, shows differences that are generally not large nor inconsistent with our overall understanding of stratospheric chemical mechanisms. Such comparisons enhance our confidence that the assessment is not subject to error from unknown chemical processes

Further improvements in stratospheric chemistry uncertainty will result from SOLVE. Preliminary results from the Photochemistry of Ozone Loss in the Arctic Region in Summer (POLARIS) mission indicate that our laboratory-derived understanding of the rate of exchange between reactive nitrogen, NOx (=NO+NO2), and less reactive reservoir compounds is somewhat inaccurate in a direction that would tend to make the atmosphere more sensitive to nitrogen emissions than previously expected. That is, the modeled ratio of NOx to total reactive nitrogen (NOy) is lower than observed (R. S. Gao et al., in preparation, 1998). Resolution of this problem will be accomplished through continued analysis of POLARIS data supplemented by new data from SOLVE along with laboratory kinetics measurements. Specifically, partitioning between NOx and HNO3 (the most abundant component of NOy in the lower stratosphere) will be examined directly with a new instrument for measuring HNO3 from the NASA ER-2. We propose that AEAP support analysis of the new instrument and data, along with the entire ER-2 measurement set, to examine the sensitivity of HSCT assessment calculations to changes in the formulation of NOx/HNO3 partitioning. This analysis will extend to comparison with corresponding measurements from the DC-8 flying at generally lower altitudes in the lower stratosphere and upper troposphere where air may be exposed to NOx emissions from aircraft in commercial flight tracks. AEAP support will enable deployment of DC-8 instruments for nitrogen species which are optional to the minimum SOLVE payload. The cost for this activity is approximately $750k.

Polar Processes

AESA assessments have demonstrated the importance of properly predicting the interaction of aircraft water, nitrogen oxides, and particulate with cold polar processes including polar stratospheric cloud (PSC) formation Processes occurring at cold polar temperatures in winter, including PSC formation, are very important to ozone because they initiate chlorine-catalyzed ozone destruction which is responsible for large seasonal ozone depletions (e.g., the “ozone hole”).

Understanding of how PSCs, sulfate aerosol, and gases interact to produce rapid polar ozone loss has improved greatly during recent years, partly as a result of measurements sponsored under

AESA. Numerous publications have documented this in the literature. Parameterization of these processes has been included in the GMI model and is used in most of the current assessment model calculations. However, our basic understanding of these processes is not complete and their simulation in models is difficult. Test calculations show that inclusion of these processes does significantly alter the calculated impact of HSCT emissions by increasing polar ozone loss, but the amount of loss varies between models depending on their method of implementation (Figure 3). Thus the assessment program has begun to quantify these effects, which were previously unquantified, but the uncertainty is still significant.

Quantitative diagnosis of the dependence of particle formation and growth, chlorine activation and recovery, and ozone loss rate on atmospheric temperature and the abundance of water, NOy, and aerosol is the main objective of SOLVE. These processes are highly non-linear, e.g., Figure 4. Precisely because of the nonlinearity of the system and the threshold behavior of the processes, small increments in NOy, H2O, or aerosol introduced by HSCT emissions are a significant concern in the polar regions. A high degree of accuracy is demanded of the modeling of these processes.

We propose that AEAP make a major contribution to the aircraft measurements of these processes. This could be accomplished by augmenting the experimental flight time of both the DC-8 and ER-2 payloads to spend more time in PSC regions and thereby increasing the chances of obtaining definitive data. Note that sampling in PSCs requires specific meteorological conditions which are often difficult to obtain within the operational limits of the aircraft. More flight time dedicated to this objective improves the probability of getting the key data. Increasing the experimental capacity by utilizing an additional aircraft, e.g., the WB-57 with a PSC-targeted payload, may also be possible. The cost of additional capacity would be about $700k. The data would provide direction for more accurate modeling of multi-phase processes in polar regions and the potential for perturbation by HSCT emissions.

Aircraft Particle Production

The impact of HSCT emissions on sulfate particles and the resultant effect on chemistry and ozone has emerged as perhaps the most significant influence of aircraft in the stratosphere. Beginning with particle measurements in the near-field plume of the Concorde aircraft in AESA Phase I, aircraft production of particles has become a major concern and uncertainty [Fahey et al., 1995]. Multi-phase reactions on sulfate particles strongly influence the balance among the main chemical ozone loss pathways in the lower stratosphere globally. AESA, subsonic, and other aircraft

research has shown that jet engines produce many more small volatile particles than expected. The mechanism and control of this production are currently not understood. The resulting atmospheric implications are potentially large, as demonstrated by AESA-sponsored modeling calculations [Weisenstein et al., 1998]. Progress in understanding this phenomenon follows from studying the process in the engine components, through the aircraft near-field, and out to global scales. Particle production has been shown to depend strongly on fuel sulfur, but the particle emission yield is still very uncertain. Model calculations have been done to test the atmospheric sensitivity of a range of particle emissions under differing atmospheric aerosol loadings, which are mainly controlled by volcanic eruptions. The range of these sensitivity tests includes impacts larger than those attributed to nitrogen oxides or water [Kawa et al., 1998].

Although particle production by aircraft is not an objective of SOLVE, the aircraft payloads and deployment regions for SOLVE do present the opportunity for gathering important data on this problem. The DC-8 will likely be deployed, at least for part of the mission, in Keflavik, Iceland which puts sampling in the commercial air traffic lanes easily within reach. Moreover, the flight lanes in winter will probably be at altitudes in the stratosphere. This forms a unique chance to study the impact of recently emitted aircraft particles (and other species, esp. NOx as discussed above) in the stratosphere. We propose to augment the basic SOLVE mission to allow the DC-8 to spend flight time explicitly in the commercial flight regions. In addition, we will support instrumentation for measuring particle size spectra to very small sizes which are emitted by aircraft. The cost for these additions is about $550k. With this data we expect to better constrain the large-scale impact of HSCT particle emission on stratospheric aerosol microphysics and chemistry.

Summary

The assessment of the atmospheric effects of high speed civil transport aircraft stands to benefit tremendously from partnership on the SOLVE mission with its primary sponsors in the Office of Earth Science (OES). The SOLVE science objectives are closely aligned with those of AEAP and the mission has been designed with cooperation in mind. We request that the Office of Aeronautics and Space Science Technology support AEAP participation in SOLVE with a budget increase totaling $3M. Distribution and expenditure of the funding would proceed through existing AEAP structure. Awards would be made in collaboration with OES via a current NASA Research Announcement (NRA-97-OES-08).

The AEAP augmentation to SOLVE would permit a much stronger experimental focus on HSCT assessment-related objectives. In combination with the basic SOLVE experimentation, compelling

new insight into the effects of aircraft emissions at high latitudes in the stratosphere is expected to result. Specifically, AEAP will support:

- balloon tracer flights and ER-2 tracer instruments to provide high latitude transport diagnostics
- ER-2 and DC-8 instrumentation for reactive nitrogen measurement to resolve discrepancies in model nitrogen partitioning
- aircraft flight opportunities to sample in regions subject to multi-phase (e.g., PSC) processes to better constrain modeling of these processes
- measurement flights into air traffic corridors and instruments for small particle measurements to examine the impact of aircraft exhaust emission in the stratosphere

These data will improve our understanding of basic processes, increase confidence in predicting the effects of HSCTs, and decrease the uncertainty in assessing the impact of HSCTs on stratospheric ozone.

References

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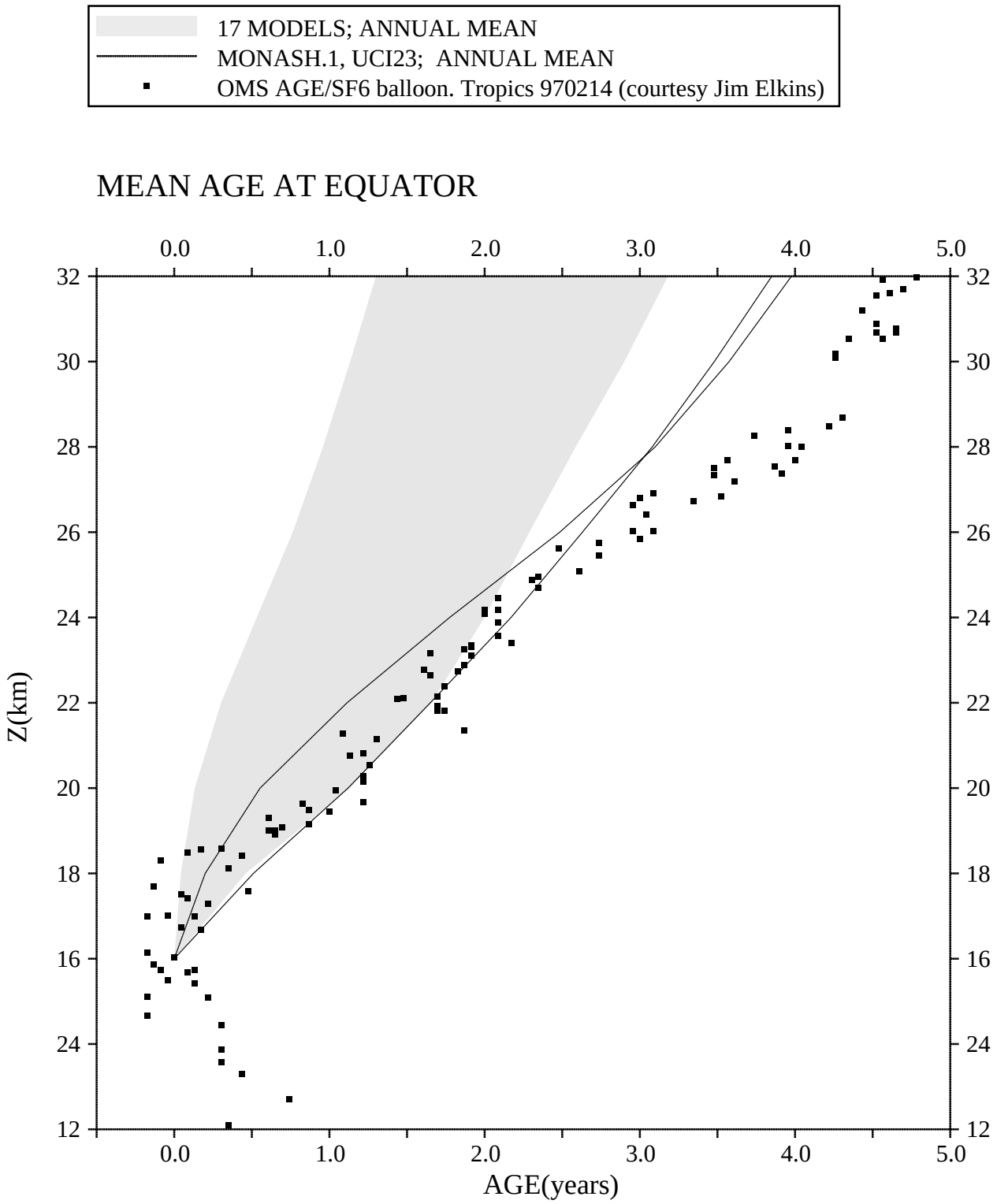
Figure Captions

Figure 1. Age of air comparison. Profiles of mean age of air are derived from measurements (dots) and models (shaded) in the tropics. Range of 17 models from M&M II is broad and well below measurements at most altitudes. Also shown separately (lines) are results from two models which come closest to the measurements in the middle stratosphere.

Figure 2. HSCT tracer dispersion. Zonal mean cross section of steady-state accumulation of aircraft NOy tracer (ppbv) is shown from several 2-D models from M&M II. Models show significant differences in tracer amount. All models used same HSCT NOy emission rate and location.

Figure 3. Seasonal and latitudinal distribution of HSCT column ozone perturbation. Set of models from AESA 98 assessment show variability of distribution of ozone perturbation for standard HSCT emission scenario not including particle emission. Note sensitivity of some models in polar regions (e.g., AER, UNIVAQ).

Figure 4. Response of reactive chlorine to temperature in multi-phase reactions. Ozone-destroying ClOx is produced rapidly from reservoir HCl (and ClONO2 not shown) as temperature decreases below apparent threshold near 195 K. Small increases in water vapor, such as might result from HSCTs, will shift the activation threshold to higher temperatures. Adapted from Kawa et al., [1997].


17 MODELS; ANNUAL MEAN
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OMS AGE/SF6 balloon. Tropics 970214 (courtesy Jim Elkins)
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Figure 1

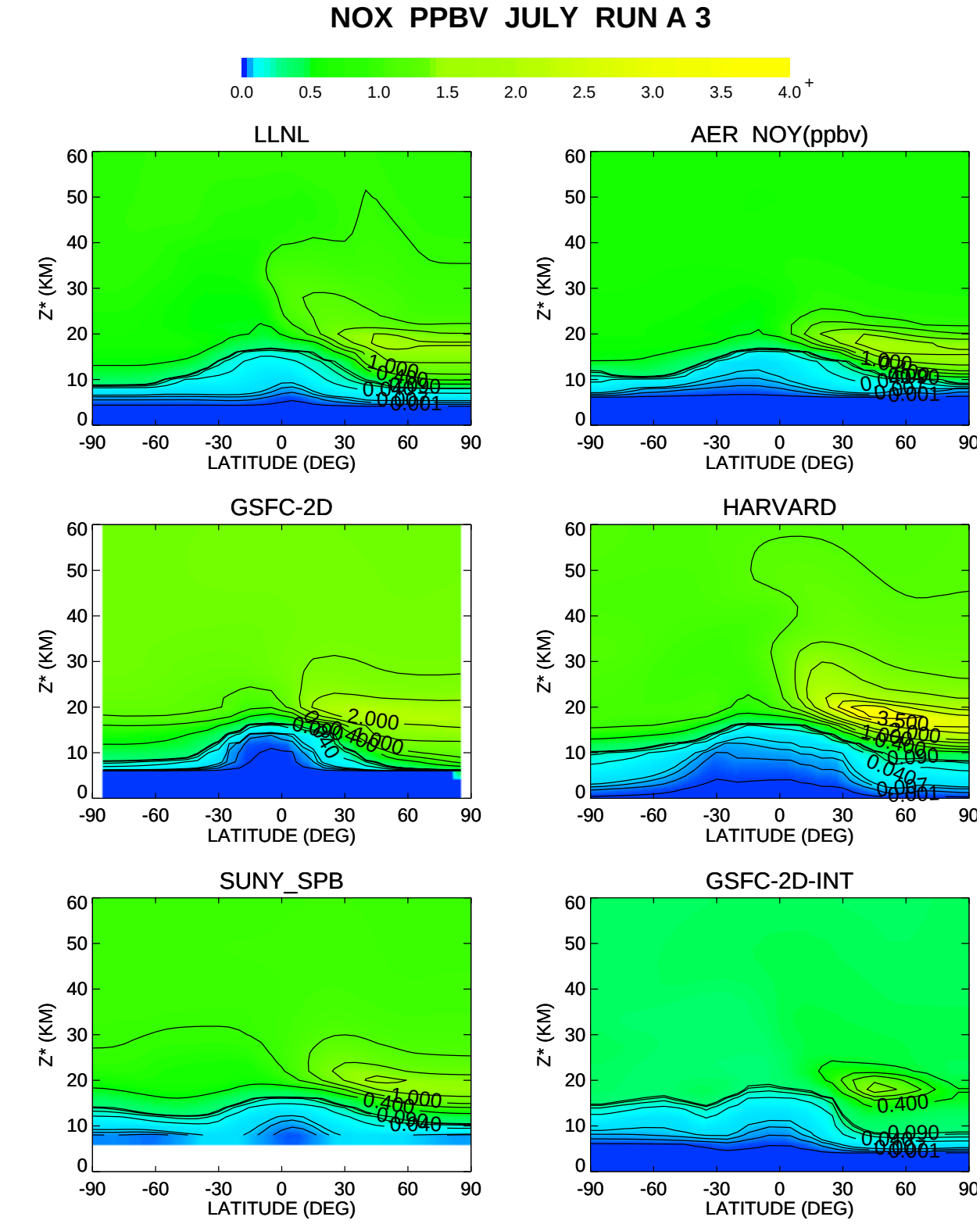
NOX PPBV JULY RUN A 3
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Figure 2

O3 Column Change (%), Scenario 4 - Scenario 1

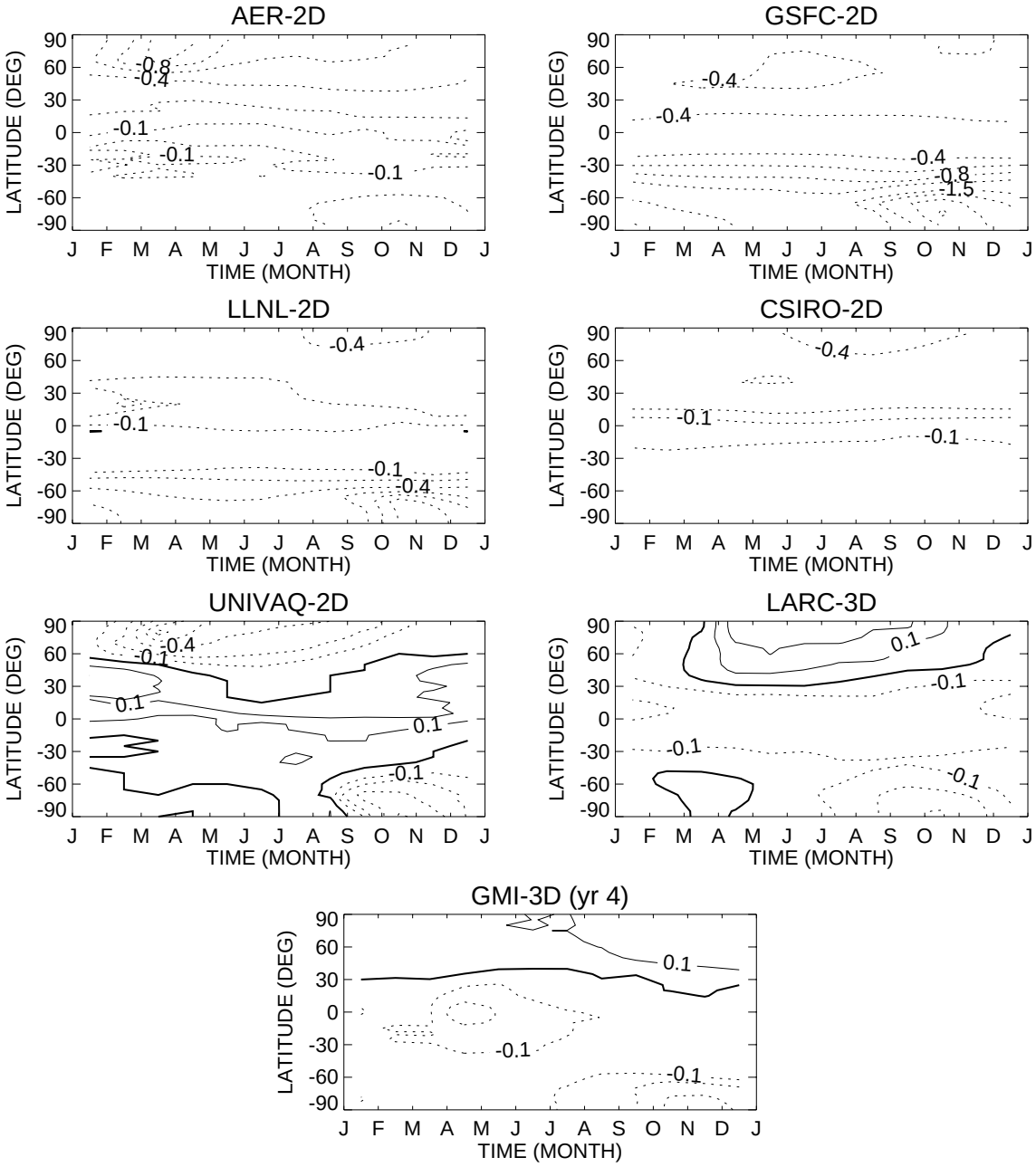

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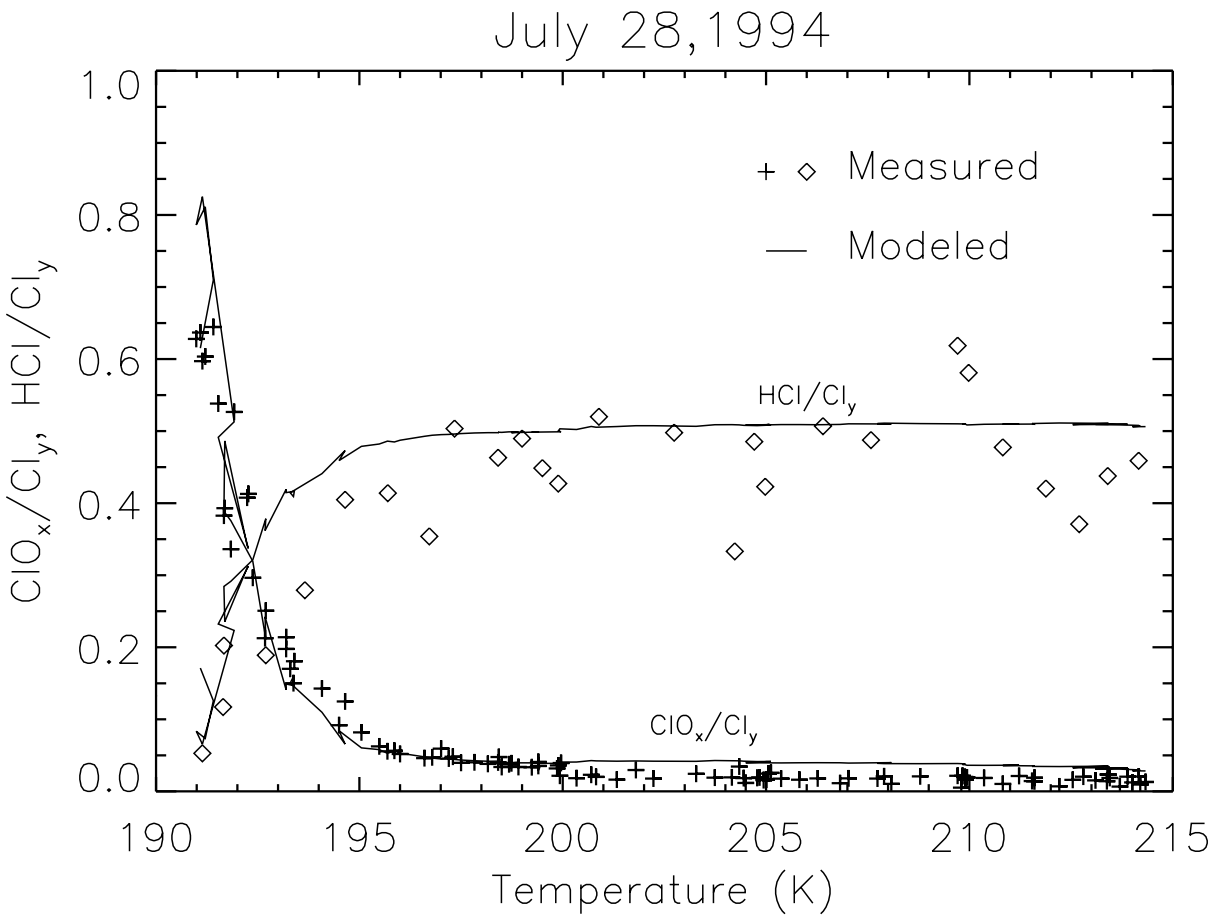
Fig 3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0.4
0.2
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ClO_x/Cl_y, HCl/Cl_y
Measured
Modeled
HCl/Cl_y
ClO_x/Cl_y
190
195
200
205
210
215
Temperature (K)

Figure 4