Chapter 6

Lower Stratospheric Measurement Issues
Workshop Report

Arthur L. Schmeltekopf
National Oceanic and Atmospheric Administration, Retired
INTRODUCTION

The "Lower Stratospheric Measurement Issues" workshop was held at NASA Ames Research Center on 17-19 October 1990. The 3-day workshop was sponsored by the Atmospheric Effects of Stratospheric Aircraft (AESA) component of the High-Speed Research Program (HSRP). Its purpose was to provide a scientific forum for addressing specific issues regarding chemistry and transport in the lower stratosphere, for which measurements are essential to an assessment of the environmental impact of a projected fleet of high-speed civil transports (HSCTs).

The objective of the workshop was to obtain vigorous and critical review of:

- atmospheric measurements needed for the assessment,
- present capability for making those measurements, and
- areas in instrumentation or platform development essential to making the measurements.

This information was obtained from experts in related fields and investigators directly involved in NASA's stratospheric measurement and instrumentation development programs. The final goal of the workshop was to develop recommendations for further study, including measurements, instrumentation, and platforms needed, in support of a focused aircraft campaign to be flown in the 1992-1994 time frame that would address critical elements in the assessment of HSCTs.

WORKSHOP RECOMMENDATIONS

Areas for further study that were stressed most during the workshop include the following:

- **We need critical tests of our understanding of the dynamics of the lower stratosphere.** These tests can best be achieved by obtaining measurements over a climatological range of tracers in the lower stratosphere (minimally: 10 to 25 km, four seasons, and at least five latitudinal locations). For that we can use both the existing data sets and obtain new, more complete data sets particularly for CO₂, CFC11, CFC12, CH₄, and N₂O. Some new instruments will be required.

- **We need critical tests of our understanding of the chemistry of the lower stratosphere.** These tests can best be achieved by obtaining a climatology of as many members of the free radical families (H, N, Cl, and O) as possible. Measurements of chemical species that currently are unmeasured are crucial for these tests. Particularly important are chemical species in the odd hydrogen, reactive nitrogen, and chlorine families.

- **We need critical tests of our understanding of radiation in the lower stratosphere.** These tests are needed to check on photolysis rates as well as hybrid dynamics (scattering). Particular attention must be paid to the possibility that clusters might be playing a role in determining the absorption spectrum (and thus photolysis spectrum) of many molecules under lower stratospheric conditions.

- **We need substantial improvement in our understanding of the character of the aerosols that exist in the lower stratosphere as well as how they might affect the chemistry and radiation there.** It is crucial to know
whether the H_2O from the engine exhaust could increase the production of H_2O-
NAT (nitric acid trihydrate) aerosols and thereby dramatically affect the chemistry in
the lower stratosphere.

- **We badly need a new aircraft platform that will get us to at least 25 km; and in addition, the new platform must not have the severe safety constraints that are imposed on the ER-2, the current principal aircraft platform.** For the studies indicated above, even if we had the proper measurement instrumentation, we would still lack a platform that can get the instruments to the right place in the lower stratosphere at the right time.

- **We must verify that the measurement techniques used by the engine manufacturers for the emission index (EI) of engines are being made using the same calibration procedures as those used in atmospheric measurements.**

- **We need to do model studies of the aircraft plumes to see whether there are any possible conditions in which plume processing could have a global impact.**

**WORKSHOP STRUCTURE**

The workshop was organized into seven sessions each of which was keyed to specific issues.

- Known Problems in Lower Stratospheric Chemistry
- Known Problems in Lower Stratospheric Transport
- What Platforms Do We Have or Can We Get
- Present Measurement Capabilities
- New Measurement Techniques
- What Do We Know about Engine Exhaust
- What Is Our Plan of Action

Each session included opening remarks from its chairman, presentations or tutorials given by invited experts, and a panel discussion. Questions were posed to panel members in advance of the workshop to stimulate critical examination of each session’s subject matter. The names of contributors and the topics they addressed, as well as the major questions asked of panel members, are given in the workshop agenda in the Appendix at the end of this chapter.

**SYNOPSIS OF SESSIONS**

Session chairmen provided a written summary of their sessions; each synopsis that follows is drawn from those summations.
SESSION 1. KNOWN PROBLEMS IN LOWER STRATOSPHERIC CHEMISTRY

Chairman

Dr. Carleton J. Howard, Aeronomy Lab, National Oceanic and Atmospheric Administration

Questions/Issues

- What critical laboratory measurements are needed?
- What critical atmospheric measurements are needed?
- Can in situ measurements help us to determine rates directly?

Synopsis

Since it is difficult to discuss in narrative form, the various ways that our understanding of the chemistry of the lower stratosphere needs to be amplified, the following section simply states and lists what is needed. For the reactions listed, the new information required is indicated in the preceding text or in comments given after the reaction. All of these measurements should be done under ambient conditions of the lower stratosphere; that is, with the temperature down to 190 K, pressure between 15 and 300 mb, and ambient mixing ratios of N₂, O₂, H₂O, etc.

Homogeneous Chemistry

Improved laboratory data are required for the following reactions under lower stratospheric conditions:

- O(1D) + N₂O  (NO product yield)
- NO₂ + NO₃ → N₂O₅
- O + NO₂
- HO₂ + NO₂ + M
- OH + HO₂NO₂  (including product yield)
- HO₂ + OH
- HO₂ + O₃
- HO₂ + NO

If HSCT engines introduce significant amounts of hydrocarbon material, then the oxidation reactions of hydrocarbon exhaust products (See item 5, paragraph 2 below) need to be measured.

Improved photolysis rates under lower stratospheric ambient conditions are needed for the following species:

- O₂  In situ measurement, preferred, including O[3P] product
- NO₂
- NO₃
- N₂O₅
- HNO₃
- HO₂NO₂
- Organic nitrates Including peroxyacetyl nitrate (PAN) (See item 5, paragraph 2)
Heterogeneous Chemistry

We need more information about the shape, size, and bulk composition of stratospheric particles, as well as the composition and morphology of the surfaces of stratospheric particles. How much and what kind of surfaces are available?

Laboratory measurements of reactions, processes, and surface accommodation efficiencies should be made using surfaces and gaseous concentrations characteristic of the atmosphere. The possibility that reactivity on surfaces is dependent upon the microstructure (the amount of roughness and the presence of cracks and fissures) should be investigated.

Phase diagrams, thermodynamics parameters, and kinetics parameters, such as growth and desorption rates, are needed for the types of particles that exist in the stratosphere.

Studies should be made of the particles emitted by HSCT aircraft, notably soot and metal oxide particles, to determine their potential roles as reaction sites and condensation nuclei in the stratosphere. Studies should be made of the affects on the photolysis rates for molecules (particularly HNO₃) tied up on particulates. What can we say about photolysis of NAT particulates?

Chemical Modeling

Modeling studies should be made to determine the kinetics and photochemical parameters that contribute the greatest uncertainties to the evaluation of HSCT environmental effects.

Model studies should be made to assess possible effects of heterogeneous processes in the stratosphere. Can HSCT emissions cause surface chemistry to play a role at lower latitudes or over longer seasons? The possible effects on halogen chemistry should be included in the evaluation.

Plume Modeling

The role of the aircraft wake vortices in entrainment and transport of exhaust material should be assessed. How much plume material is dispersed into the stratosphere? The possibility of significant chemical processes occurring in the exhaust plume should be assessed. For example, is the NOₓ emission conversion to HNO₃ enhanced in the plume?

Miscellaneous Topics

High-quality quantum mechanical studies should be made of selected critical reactions whose rate coefficients, temperature, and pressure behavior are poorly understood, for example, HO₂ + OH and HO₂ + O₃. Similar studies of reactions that occur on or in particles, such as HCl + ClONO₂ → Cl₂ + HNO₃, would be very valuable.

Accurate data are needed on the emissions from proposed HSCT aircraft engines operating at ambient conditions. Nitrogen oxide, hydrocarbon, water vapor, and particle emissions are the most important factors.

Stratospheric perturbations resulting from the current subsonic aircraft fleet should be evaluated. Possible effects from both nitrogen oxide and sulfur oxide emissions should be improved. The amount of NOₓ contributed to the stratosphere by lightning should be assessed as well.
SESSION 2. KNOWN PROBLEMS IN LOWER STRATOSPHERIC TRANSPORT

Chairman

Dr. James R. Holton, Department of Atmospheric Science, University of Washington

Questions/Issues

- Will SST exhaust really mix upward?
- Can the high-resolution structure of the transport be important?
- Can exotic tracer experiments be useful?

Synopsis

The discussion of constituent transport can be divided into three categories: (1) the climatology of global transport, particularly as it influences the global distribution of pollutants emitted by aircraft; (2) mechanisms of transport in the lower stratosphere, particularly in regard to stratosphere-troposphere exchange rates at all latitudes and horizontal transport rates in the polar regions; and (3) behavior of aircraft plumes from the wake vortex breakup stage to large-scale dispersion. Issues in each of these categories are discussed next.

Global Transport Issues

The climatology of NOx is important in evaluating "traditional" aircraft-related ozone depletion concerns (i.e., homogeneous NOx chemistry). NOx enhancements above an altitude of ~15 km are expected to deplete ozone, while below that altitude, smog chemistry is expected to produce ozone (neglecting possible complications of heterogeneous reactions; see Figure 1). The major transport issue for this piece of the total problem is to determine the climatology (i.e., the latitude and height distribution and the seasonal variability) of the steady-state perturbation to the "natural" NOx distribution resulting from emissions from a fleet of HSCTs. There is general agreement that the magnitude of this effect, and its influence on the ozone layer, will be strongly dependent on the altitude at which the HSCTs fly. But, whether there is a "safe" altitude will depend (among other things) on the fate of the emissions introduced into flight corridors. There are important questions concerning the extent to which emissions occurring at a given flight altitude might disperse to higher altitudes, where the ozone depletion potential is greater. If there is rapid quasi-isentropic dispersion from the Atlantic and Pacific flight corridors into the tropics, then emissions would enter the tropical upwelling region and be advected to higher altitudes. A direct observational proof that molecules emitted in the proposed flight corridors will penetrate to any given altitude and latitude would apparently require continuous emission of an exotic tracer into a flight corridor and its concentration measured at a wide range of latitudes and heights for a year or more. The workshop participants, by and large, agreed that there was no practical method to accomplish such a direct test of large-scale plume dispersion. Thus, the only viable way to predict the likely global perturbation of the NOx distribution by a fleet of HSCTs is by using models that are testing by a wide variety of observations.

Although not all agree, the consensus view is that this type of tracer dispersion study cannot be done in the framework of two-dimensional (2-D) modeling. Eddy mixing by planetary wavebreaking and synoptic scale motions differs considerably from eddy diffusion. There is a strong up-down asymmetry in large-scale mixing processes that cannot be modeled in terms of eddy diffusion. For example, 2-D models seem incapable of simulating the transport into the troposphere that is associated with tropopause fold events.
Studies of global tracer transport must be based on three-dimensional General Circulation Models (GCMs). The basic requirement for such models is that they explicitly resolve the scales that are important for global transport so that parameterization effects are minimized. This implies high vertical resolution (1 km or better, perhaps as low as a few hundred meters) near the tropopause, and sufficient horizontal resolution (a few hundred kilometers at most, and perhaps as few as 10 km) to accurately represent the important scales of eddy variability in the lower stratosphere. Model validation should utilize all available data sources. These include global satellite data (particularly Upper Atmosphere Research Satellite [UARS] trace constituent data), balloons, ground-based remote sensing, and aircraft data. Model validation should also utilize $^{14}$C and other radionuclide data from the nuclear weapons testing era. If a model that is calibrated on the basis of one set of tracer data can simulate the global distribution and seasonal variability of all other independent tracers, it may be a credible tool for evaluating the global distribution of pollutants emitted by a fleet of HSCTs. Such a model could then be used to improve the eddy diffusion parameterizations in the 2-D models that will be required for assessment studies.

Figure 1. Ozone formation from the smog reactions based on methane and nitrogen oxides (45° latitude, spring).
Transport Problems Related to Heterogeneous Ozone Depletion

Heterogeneous chemical processes are among the most challenging of the problems facing the HSRP. Aircraft engines are not only sources of NO\textsubscript{x} but of water vapor, particulates, SO\textsubscript{2}, and perhaps other trace gases as well. Perturbations in these constituents may affect, and be affected by, polar stratospheric clouds (PSCs). Modeling studies at GSFC show, for example, that for the Seattle-London polar flight route an HSCT would fly within the winter polar vortex quite regularly (although this route would be over land, and therefore, might not be allowed at supersonic). If patches of air with elevated levels of water vapor, particulates, and other emissions remain coherent for sufficiently long periods and encounter sufficiently low temperatures, heterogeneous processes leading to ozone depletion might be activated. This possibility must be explored not only for the polar regions, but also near the tropical tropopause, which is also a very cold region that would be affected by any HSCT flights to the southern hemisphere. Investigations of this class of problems cannot be done with UARS or other planned satellite data. UARS trace constituent data will have poor coverage below 20 km, and poleward of 78° latitude. The data also will suffer degradation if there are clouds in the field of view. High vertical and horizontal spatial sampling is needed in the 15-25 km altitude range at a wide number of latitudes to evaluate the potential for heterogeneous processes. Emissions of water vapor by an HSCT fleet may prove to be just as significant as NO\textsubscript{x} emissions, and data are needed to define the current water vapor climatology as well as to estimate HSCT impacts. The objective must be more than just process studies of PSCs; changes resulting from emissions of water vapor and aerosols should be assessed as well. Aircraft and balloon observations at a number of latitudes and seasons are required.

Small-Scale Dispersion of Engine Emissions

Dispersion from the wake vortex breakup stage (scale of hundreds of meters) to the large scale (hundreds of kilometers) is not well understood. Deformation by vertical shear of the horizontal winds appears to be the most important process for dispersion on these scales. It is likely that vertical dispersion will be much slower than horizontal dispersion because of the influence of vertical shear. Since the mean winds have a strong seasonal dependence, the rate of dispersion from mean wind shear may also have a strong seasonal dependence. This range of scales is too small to be explicitly resolved in global 3-D models, and so must be parameterized. Thus, more information on dispersion from the small scale to the GCM resolved scales would be highly valuable as an aid to improving tracer simulations based on 3-D models. Direct observations of the plume dispersion in this scale range would clearly be very difficult. Exotic tracers might be useful for this scale, but no specific experimental strategy was agreed on in the workshop.

SESSION 3. WHAT PLATFORMS DO WE HAVE OR CAN WE GET?

Chairman

Dr. Art Schmeltekopf, National Oceanic and Atmospheric Administration, Retired

Questions/Issues

- Do any of these platforms get us far enough?
- High enough?
- Long enough (duration)?
• When can the platforms be ready?

Synopsis

Four types of platforms are available to the HSRP for making the measurements needed to demonstrate our understanding of the atmosphere: 1) satellite, 2) balloon, 3) aircraft, and 4) ground based. The science problems faced by the HSRP are mostly in the 12 to 25 km range and require that measurements be made in many locations around the globe, in all seasons. These measurements are required to be made with high spatial resolution. They need to be made in all types of weather conditions: cloudy, in and out of jet streams, in the polar night, etc. Because the satellite and ground-based platforms are long path and require good visibility, they cannot yield all of the information needed. Balloons are very hard to position accurately, so it would be unlikely that measurement at an exact location in a jet stream could be made, at least on a routine basis. However, the high-resolution vertical soundings that can be made by balloon will provide useful data when available. It is clear that the measurements made by UARS will provide a very important part of the global climatology for many of the species of interest; however, most of the measurements cannot be made low enough, in the dark, or in or below the cloud decks. Ground-based high-resolution lidar and other instruments will provide a very good check on the satellite and aircraft measurements, but they cannot be made in every location and are not possible under cloudy conditions.

It is therefore clear that at least some of the measurements critical to the HSRP mission must be made from an aircraft platform. There are five aircraft platforms available now or proposed for the near future. They are: ER-2, Condor, Perseus, Theseus, and the High-Altitude Aircraft Research Program (HAARP). In addition there are two supersonic platforms that could be used: SR-71 and the Concorde.

The idea of using a supersonic platform was suggested; the obvious problem—Mach heating in the sampling process—was given as the reason that supersonic platforms are not being considered for use by the HSRP. Two ideas were presented to avoid the Mach heating, but both involved cooling the stream through interaction with a cool wall. The effect of the wall on the reactive species that are to be measured was considered prohibitive. In addition, no proposed method would allow the faithful sampling of aerosols from a supersonic platform. The Concorde representatives offered the possibility of using the Concorde as a sampling platform, and that may be feasible for the stable species.

For the reasons stated previously, the most desirable platform for the HSRP is the subsonic aircraft platform, and so the major discussions in the platform session focused on them. The major comparisons to be made about our choices for the subsonic aircraft platform are in Table 1.

ER-2 Platform

The ER-2 needs no discussion since it has been the workhorse used for measurements in the lower stratosphere for many years. For this program has four major limitations: 1) a maximum altitude of ≈20 km, at least 5 km below our needs; 2) a range of 5400 km, which will not allow us to get to many important areas from usable airports; 3) since it has a single engine and a pilot, safety requirements place too many restrictions on where and when it can be flown; and 4) it does not operate well below 16 km, and so is not suitable for making many measurements below that altitude (obviously it passes through the altitudes below 16 km on takeoff and landing) and therefore it cannot meet the full requirements of HSRP mission. Although several improvements, are planned for the aircraft, important for its present community of users, they will not substantially affect the limitations stated above.
<table>
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<tr>
<th></th>
<th>ER-2</th>
<th>Condor</th>
<th>HAARP</th>
<th>Perseus Gasoline</th>
<th>Perseus Hydrogen</th>
<th>Theseus</th>
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<td>Mod turb</td>
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* Pilot on board/remotely piloted.

NOTES: The ER-2 is powered by a turbojet, while the others are propeller driven. The ER-2 cruise Mach number does not vary with altitude, but for the others it (and airspeed) varies with the cruise altitude. Fuel consumption is a function of altitude so that endurance and range are strong functions of altitude and weaker functions of payload weight. The distance that can be covered is a weaker function of payload weight and altitude. Since the propeller-driven aircraft have a much lower air speed (a very strong function of altitude), the atmospheric winds have a larger influence on the distance traveled than it does on the ER-2. The ER-2 is a manned aircraft, the others are unmanned. The HAARP will be built for either manned or unmanned flight: manned for testing and unmanned for long missions. The Condor is a twin-engine aircraft, the others are single engined. The HAARP is towed off the ground by a Twin Otter or equivalent. "Any" runway surface means paved, grass, dirt, snow, or ice.
**Condor Platform**

The Condor was designed as a station-keeping, high-altitude platform. Since it is an unmanned, long-duration, twin-engined aircraft, three of the problems associated with the ER-2 are overcome. Its only disadvantage is that its maximum altitude is essentially the same as that of the ER-2. The aircraft has had several successful test flights; however, it is not mission-ready and will need further development before it is useful for HSRP.

**Perseus Platform**

The Perseus is a highly specialized aircraft. It is expected to be able to take small, light experiments to near 30 km. It now (November 1990) exists as an untested airframe with an as yet untested engine. There seems little doubt that the concept behind it is sound and that the aircraft is likely to perform as expected, but currently it now has several weaknesses. It can remain at altitude for only very short periods and has limited range. On the other hand, it can be launched from almost any solid surface and can thus be taken to most places of interest and launched there. Another weakness is the fact that its payload-carrying capacity of about 50 kg (analogous to a jeep) is so low that it is difficult to get many experiments on board for simultaneous measurements. The proponents of this aircraft expect that the cost per copy of the aircraft is so low that several could be flown at once for simultaneous measurements.

**Theseus Platform**

The Theseus design concept is expected to overcome several of the problems associated with Perseus. It is expected to carry a heavier payload (analogous to a van) and fly much farther. As now proposed, however, it could not be launched from just any solid surface, but would require a runway.

**HAARP Platform**

The HAARP is a design concept developed at the request of the NASA Ames Research Center to meet the specifications determined at a workshop held in Truckee, CA, July 15-16, 1989. This aircraft is designed to overcome all of the weaknesses that are evident, in some way, in all of the other aircraft. Thus the aircraft resembles a truck because of its payload capability. It is fairly expensive and it will take several years before it reaches the operational stage (maybe too long to be of interest to the HSRP). It clearly would be a very useful platform to all of the atmospheric science community.

**Conclusion**

The real difficulty with the platforms described previously is that the atmospheric science community has no way of generating financial support for all of them. The community will thus have to formulate a consensus view and support at most one or two of these platforms. We have the ER-2, so it is clearly a player for the foreseeable future. The Condor does not get high enough to fulfill all of the needs of HSRP. Since the ER-2 can do much of what the Condor does, the Condor cannot be the aircraft of choice. The clear long-term choice is an aircraft that will follow the HAARP's design, but this aircraft cannot be ready for several years and this program needs results before then. It seems clear that the Perseus is the only new platform that has a chance of getting the measurements needed above 20 km in the time frame allotted for the HSRP.

There have been strong advocates for the three new platform designs: Condor, Perseus/Theseus, and HAARP. During the HSRP workshop at ARC those groups got together
to form a loose team and try to help one another as much as possible. Having realized that Perseus is a very viable platform that could solve not only some of our atmospheric measurement issues but also test the models used for calculating the wing, propeller, and engine cooling designs (major areas of concern for the design of Theseus and HAARP), these advocates have now agreed to assist one another in any practical way, sharing information, etc.

SESSION 4. PRESENT MEASUREMENT CAPABILITIES

Chairman

Dr. James G. Anderson, Engineering Sciences Laboratory, Harvard University

Questions/Issues

- Are all of our present capabilities accurate enough?
- Specific enough?
- Fast enough?
- Do we currently have the "Right Stuff?"

Synopsis

Instrumentation available for the HSRP comes from a lineage of balloon and aircraft-borne developments encompassing the past 10 or more years. In Tables 2, 3, and 4 plus the sections that follow, we review the current state of instrument technology with the following constraints:

- The HSRP program needs place maximum emphasis on the dynamics, radiation, and chemistry in the 10 to 30 km region. Thus, any instrumentation for this program must have good spatial and temporal resolution with signal-to-noise ratios of 10 or greater throughout the altitude interval.

- There is an ongoing Upper Atmosphere Research Program (UARP) that supports field efforts (e.g., large-lift, high-altitude balloon soundings) that are too extensive to describe here fully. We will abstract only those instruments that are directly applicable to the needs of HSRP.

- Instruments must be identified with the platforms on which they are deployed; otherwise, an explicit deployment strategy is difficult to articulate.

The challenge for HSRP is to craft a collection of instrument/platform combinations that can push the field forward in the 1992 time frame and stage a new combination of dynamics, chemistry, and radiation observations for 1993-94 which will diagnose for the first time the mix of dynamics and chemistry so central to the scientific objectives of HSRP. For example, the ER-2 instrument array emulates the measurement combination that must be deployed from Equator to pole with intensive uninterrupted coverage in the 10-km to 30-km altitude interval to sort out superposition of transport and chemistry. That platform can contribute to three sub-campaigns:
- Polar Campaigns - to construct a picture of the stratosphere up to 20 km north of 30° N latitude and south of 30° S latitude.

- Tropical Campaigns - to define the dynamics and photochemistry of the region for 30° S to 30° N latitude. This mission is especially important because we have so little in situ information for this region.

- Tropopause Campaigns - to lay the foundation for understanding stratospheric/tropospheric exchange. We need a new approach here, defining some new ideas about how the process works. The Stratosphere-Troposphere Exchange Project (STEP) campaign was focused on what, apparently, is not the most important process, so we need some new ideas.
Table 3. Current Instrument Capabilities Part II: Balloon-Borne, In Situ and Remote

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Technique</th>
<th>Altitude</th>
<th>Detection</th>
<th>PI/Institution</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO, NO₂</td>
<td>TDL/BLISS</td>
<td>23-35 km</td>
<td>150 pptv for NOₓ, NOᵧ</td>
<td>Webster/JPL</td>
<td>Heavy lift balloon</td>
</tr>
<tr>
<td>HNO₃, O₃</td>
<td></td>
<td>Balloon restric. limit altitude</td>
<td>NOₓ</td>
<td>JPL</td>
<td></td>
</tr>
<tr>
<td>CH₄, N₂O</td>
<td>LIF, resonance</td>
<td>23-38 km</td>
<td>1 pptv OH/HO₂</td>
<td>Anderson/Harvard</td>
<td>Heavy lift balloon</td>
</tr>
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<td>H₂O, ClO,</td>
<td>Fluorescence,</td>
<td>23-38 km</td>
<td>100 pptb H₂O</td>
<td>JPL</td>
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<tr>
<td>BrO, O₃</td>
<td>UV absorption</td>
<td>Balloon restric. limit altitude</td>
<td>2 pptv ClO, BrO</td>
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<tr>
<td>NO, NOᵧ</td>
<td>Chemilumin.</td>
<td>23-38 km</td>
<td>50 pptv</td>
<td>Ridley/NCAR</td>
<td>Part of Harvard gondola</td>
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<td>HCl, HF,</td>
<td>Far infrared</td>
<td>25-40 km</td>
<td>Remote sensing S/N good above 30 km</td>
<td>Traub/CFA</td>
<td>Heavy lift remote sensing</td>
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<td>HOCl, NO₂,</td>
<td>FIRS-2</td>
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<td>O₃</td>
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<td>O₉, NO₉</td>
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<td>25-40 km</td>
<td>Remote sensing</td>
<td>Toon/JPL</td>
<td>Heavy lift balloon</td>
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<tr>
<td>NO₉, HNO₉</td>
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<td></td>
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<tr>
<td>HCl, ClONO₂</td>
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<tr>
<td>ClO, O₉</td>
<td>Microwave,</td>
<td>25-45 km</td>
<td>100 pptv</td>
<td>Stachnik/JPL</td>
<td>Heavy lift balloon</td>
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<td>HCl, HO₂</td>
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<td>Normal</td>
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<td>OH</td>
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<td>28-45 km</td>
<td>5 pptv</td>
<td>Pickett/JPL</td>
<td>Heavy lift balloon</td>
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Table 4. Current Instrument Capabilities Part III: Radiation Measurements, UV through IR

<table>
<thead>
<tr>
<th>MEASUREMENT</th>
<th>INSTRUMENT</th>
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</thead>
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<tr>
<td>IR radiance, difference, directionality, divergence</td>
<td>IR Interferometer</td>
</tr>
<tr>
<td>IR broadband flux, divergence, difference</td>
<td>Pyrogeometer</td>
</tr>
<tr>
<td>Visible radiance, difference, directionality, divergence</td>
<td>1/4 meter spectrometer with diode array</td>
</tr>
<tr>
<td>Visible flux, divergence, difference</td>
<td>DC pyranometer</td>
</tr>
<tr>
<td>UV radiance, difference, directionality, divergence</td>
<td>DC 1/4 meter spectrometer with diode array</td>
</tr>
<tr>
<td>UV flux, divergence, difference</td>
<td>DC integrating radiometer</td>
</tr>
<tr>
<td>Water vapor, liquid, ice</td>
<td>Lyman-alpha FF</td>
</tr>
<tr>
<td>Ozone</td>
<td>In situ UV absorption</td>
</tr>
<tr>
<td>CO₂</td>
<td>In situ IR absorption</td>
</tr>
<tr>
<td>CCN aerosols</td>
<td>PMS ASASP-X-M</td>
</tr>
<tr>
<td>Cloud drop and aerosols</td>
<td>PMS FSSP</td>
</tr>
<tr>
<td>2-D ice crystal images</td>
<td>PMS 2DP</td>
</tr>
<tr>
<td>Temperature, pressure, relative humidity</td>
<td>Aircraft data, MMS</td>
</tr>
</tbody>
</table>
A limited number of heavy-lift balloon launches will provide an occasional glimpse of higher altitude constituent fields. This combination of observations will yield major advances in our understanding.

An attempt should be made to compare the instruments on different platforms, particularly the instruments on balloons, aircraft, and satellites. These intercomparisons, when they are done well (i.e., in the same air mass), give us confidence that the claimed accuracies are real. In the same vein, it is desirable to measure all of the members of a family of molecules that are linked together with fast photochemistry, and then compare the ratios of the members' density to that expected from the theory. From these results one can find errors in either the experiment or the theory; however, in order to close the gap, we need very-high-accuracy experiments.

The goals for HSRP leading to a possible quantification of exhaust emission impacts on global ozone must, however, derive from a careful tailoring of new instrument technology in radiation, dynamics, and chemical observations with new platform development (specifically, high-altitude aircraft), to "sew" the upper troposphere to the middle stratosphere.

Needs for the Future

Recommendations for Radiation Field Measurements in the Lower Stratosphere

Measurement of the direct and diffuse radiation fields at various altitudes in the upper troposphere and lower stratosphere from the S-R bands to the near infrared is needed. Measurements should be made over geographical regions of different, but well-defined, albedo (clouds, ocean, snow, desert, etc.). These measurements would allow detailed comparison with the various multiple scattering theories currently in use in interpreting stratospheric photochemistry. The influence of multiple scattering is large (as much as a factor of 3 in the radiation field [including the direct solar flux]). Absolute level of accuracy required is not high, but it should be at least 20%. Measurement pertinent to j(O2) is important in the S-R region. Spectral resolution should be sufficient to verify the models, which is not very high unless detailed modeling of the S-R region is desired.

Measurements of direct and diffuse radiation in and out of clouds (high cirrus and PSCs) would be highly desirable. Ancillary measurements of cloud properties would be desirable (size, density, depolarization ratio, etc.).

Measurement of the IR radiation field is needed for determining the effects on the local energy balance of clouds, albedo, and aerosols. Radiative transfer codes use the line/line spectra from the AFGL tapes and so line/line measurements would be most useful in testing and comparing for these types of models. This would require an FTIR type instrument to adequately resolve the line widths. Specific target species for the lower stratosphere could be H2O, N2O, O3, ice, CO2, and CFCs. In making the measurements for use in model comparison and calculation, it would be necessary to know the temperature profile under the aircraft as well as cloud heights. The effects of clouds and their microphysics (particle size and density) would also be important, particularly for NAT and sulfuric acid aerosols.

Tracer Observations

A glaring practical shortcoming in the measurement arsenal for HSRP is the unavailability of a lightweight instrument for the "real time" detection of tracers, specifically CFC-11, N2O, and CH4. This array, along with CO2, would provide invaluable maps of:

- tracers that fall off at dramatically different rates with altitude; and
• a tracer (CO₂) that would quantify the age of a particular air parcel.

For H₂O, we are acquiring a fairly good database, but the precision and accuracy needs some improvement. It is particularly important that we measure H₂O accurately since the equilibrium temperature at which NAT forms is a function of the H₂O concentration, and the chemistry of the lower stratosphere is a sensitive function of the occurrence of NAT. It is thus possible that the aircraft emissions of H₂O could affect the chemistry of the lower stratosphere by affecting the production of NAT aerosols.

Reactive Constituent Observations

The changes that have occurred in the last 15 years regarding the calculated predictions for O₃ depletion have resulted from changes in our understanding of the chemical links between reactive nitrogen and odd hydrogen and of the effects of heterogeneous chemistry. We now have the capability to measure most of the important reactive species (Table 4), but some notable exceptions exist. We have no measurements of the odd hydrogen radicals OH and HO₂ and the species that links chlorine and nitrogen, ClONO₂. Our capability to measure NO₂, perhaps the key radical in the lower stratosphere, has not been critically tested for aircraft instruments. We must redouble our efforts to make these measurements. Equally critical is the need to establish the climatology of these reactive constituents. The emerging construction of a climatology for NOₓ, for example, has presented us with some interesting scientific questions. Imagine what we will learn when we have acquired simultaneous climatologies for all the important radical species.

There is a clear need for more accurate aerosol measurements. At present, particles that contain sulfuric acid, nitric acid, and water in various abundances in several phases are known to play a role in the chemistry of the stratosphere. Our ability to characterize the composition of these aerosol particles must be improved in order to understand their conditions of formation and their effect on the local chemistry, especially in chemically perturbed air parcels. Expectations are that new light-scattering instruments could significantly improve the accuracy in quantifying particle concentrations and sizes in the range of ~ 0.1 to 20 micron radius. Composition information could be obtained from the particle refractive index as determined from scattering polarization. Success in interpreting new measurements of size, abundance, and composition of aerosol particles is essential to further progress in this area. More detailed information on the full composition of particles that may eventually be needed may be forthcoming near the end of the HSRP program from aerosol mass spectrometer instruments currently under development in the laboratory.

Platforms

The critical variables of altitude and range must be tailored to the scientific objectives, in combination with constraints on payload weight, instrument capability, etc. The desired circumstance, of course, would be to have at least a 300-kg payload capability to 30-km altitude with a range of 3000 to 10,000 km or greater available immediately in an unmanned aircraft platform (with true costs at or below $1000/hr for operating expenses). While this aircraft may be available in the Theseus by 1994, there is a natural evolution of technology using the ER-2 and Perseus as initial platforms (which can advance the HSRP program in the near term) culminating in a more powerful combination of platforms, including the HAARP, approximately 3 to 4 years from now.

Specifically, we envision an evolution that:
Uses the ER-2 to stage polar campaigns, tropical campaigns, and tropopause studies up to 20 km. The ER-2 will also serve as a test bed for new instruments along their natural evolutionary path to lightweight status.

Perseus will be used to deploy subsets of the full instrument array either to high altitude for short duration or for longer duration at lower altitude.

SESSION 5. NEW MEASUREMENT TECHNIQUES

Chairman

Dr. Michael J. Kurylo, National Aeronautics and Space Administration, Headquarters

Questions/Issues

- Which of these new areas looks promising enough to warrant development for use in atmospheric research?
- Can they be ready by 1992?
- By 1994?
- When?

Synopsis

The presentations made during this section of the Workshop clearly demonstrated the vital need for strong synergistic relationship between the HSRP and atmospheric research programs within NASA and other U.S. Government agencies, in order to provide the measurement capabilities for evaluating future atmospheric environmental issues, including those associated with HSCT operations.

The Workshop reports and discussions on new measurement techniques were focused primarily on aircraft-borne instrumentation. This is an area that has seen considerable growth over the last 5 years, largely as a result of developments associated with the Stratosphere Troposphere Exchange Project (STEP), the Airborne Antarctic Ozone Experiment (AAOE), and the Airborne Arctic Stratospheric Expedition (AASE) conducted in the late 1980s by NASA's Upper Atmosphere Research Program (UARP). The current suite of instruments designed for use on the NASA DC-8 and ER-2 aircraft represents a very powerful atmospheric observation capability. These existing measurement opportunities are enhanced further by the more mature balloon-borne and ground-based instrumentation deployed by the UARP for more than a decade to develop atmospheric climatologies in several latitude regions. Nevertheless, there are many recognized limitations in atmospheric sampling that are yet to be overcome. Presentations during this session of the HSRP Workshop demonstrated the wealth of scientific talent now preparing to meet these challenges. Ongoing and planned research activities, while in many cases directed at generic aspects of atmospheric measurement science and trends detection, will provide the capabilities needed for addressing the environmental issues confronting the development of a HSCT.

Presentations on both in situ and remote-sensing instruments were included in the program. While the dominant focus was on existing platforms (i.e., DC-8, ER-2, and large balloons), it was clear from the discussions that there is a growing consensus for directing a
component of instrument development activities towards deployment on small lightweight balloons and/or high-altitude (manned or unmanned) aircraft.

**Lidar Instrumentation**

Among the remote-sensing instruments, significant improvements have been realized in lidar instrumentation (funded in part by NASA's UAR and Tropospheric Chemistry Programs). Capabilities now exist for investigating background aerosols and all types of stratospheric clouds, including Type II PSCs. Aerosol scattering and depolarization measurements are presently possible (or will be available within the time frame of the HSRP) from the DC-8 in either the zenith or scanning modes and from the ER-2 in nadir, zenith, or scanning modes. Plans are under way to extend the existing DC-8 ozone lidar measurements to an altitude range of 28-30 km. While the current DC-8 water vapor lidar instrument has been limited to measurements in the troposphere, existing and developing technologies could extend such measurements to approximately 8 km above flight altitude within the next few years. An ER-2 tropospheric system currently under development could also be modified for lower stratospheric studies on a similar time scale. Lidar measurements could be used for several other important atmospheric constituents. For example, development of a Raman lidar and its deployment on the DC-8 for simultaneous measurements of stratospheric methane, water vapor, nitrogen, oxygen, and temperature has been proposed and is being evaluated.

**Microwave Instrumentation**

There have also been improvements in microwave/submillimeter limb-sounding instrumentation. A reasonably lightweight package (developed under NASA's UARP), suitable for balloon or aircraft platforms, can simultaneously measure several species (ClO, O3, HCl, HO2, HNO3, and N2O) important in atmospheric ozone chemistry and can be extended to other species through the addition of appropriate filter banks. The instrument has the best signal-to-noise ratio and resolution with the platform situated above the atmospheric layer to be measured.

**Infrared Instrumentation**

Similar advancements in infrared (IR) instrument development have also been achieved. A new tunable diode laser spectrometer has been fabricated for the ER-2 under UARP support and is about to undergo test flights. A four-channel instrument, it is designed to make simultaneous in situ measurements of NO2, HNO3, HCl, CH4, H2O, and O3, thereby adding considerably to the existing IR absorption capability on that platform. Some early proof-of-concept investigations have also been conducted for an open path IR absorption cell that should eliminate sampling problems for some of the more reactive gases and may have greater sensitivity than other instruments because the achievable path length is longer. Also under development (with HSRP support) is a laboratory proto-type instrument for in situ CO2 measurements that use non-dispersed IR analysis. Construction of a flight instrument (scheduled to begin within a year) should greatly enhance dynamic tracer measurement capabilities from the ER-2.

**Instrumentation for In Situ Measurement of Radical Species**

There has been considerable development of instruments for the in situ measurement of important radical species. Laboratory studies and instrument prototype development has been under way within the UARP for a lightweight OH/HO2 monitoring instrument. This development (based in large part on the heritage of high altitude balloon instrumentation) will be extended to a package suitable for ER-2 integration under HSRP support. NASA's UARP has also supported the development of a lightweight ClO/BrO instrument (which can be flown on a small balloon or a high-altitude aircraft) and will initiate support for development of a similar
instrument for in situ measurement of ClONO₂. The measurement of various paramagnetic gases (i.e., radicals such as Cl/ClO, OH/HO₂, NO/NO₂) may also be possible using a mid-IR magnetic rotation spectrometer. More detailed laboratory study would be required to ascertain the potential of this technique. Finally, the ER-2 chemical measurement suite will soon be further enhanced even with the HSRP-sponsored conversion of the existing NO/NOₓ instrument to a three-channel system capable of simultaneous measurements of the species.

**Instrumentation for Real-Time Atmospheric Tracer Profile Measurements**

One of the more significant gaps in chemical sampling instrumentation lies in the area of real-time atmospheric tracer profile measurements. While the previously mentioned CO₂ instrument will play an important role in this area, there is a need for a lightweight instrument package suitable for deployment on a small balloon or high-altitude aircraft and capable of recording the profiles of CFC₁₃, CF₂Cl₂, or N₂O to altitudes above the current ER-2 limit. The potential for the development of such an instrument may lie in fast-response gas chromatographic technology or ion mobility spectrometry, and is currently being evaluated.

**Instrumentation for Physical and Chemical Characterization of Condensation Nuclei and Aerosols**

There have also been (and continues to be) improvements of and developments in instrumentation for the physical and chemical characterization of condensation nuclei (CN) and aerosols. For example, sizing capabilities were greatly improved between the AAOE and AASE. While there are now several new techniques and inlets for sampling the various aerosol size ranges, in general particles over 0.2 μm in diameter must be sampled in situ, while smaller sized particles can be brought into the aircraft for analysis. The HSRP is supporting three different instrument studies in the CN/aerosol area. These include improvements in CN counting for the ER-2 instrument, laboratory studies leading to the development of a stratospheric soot-measuring instrument, and development of an aerosol mass spectrometer (laboratory prototype of an aircraft instrument). The latter should yield not only aerosol size and composition information, but also provide identification of trace species in the particles. Finally, there are a number of laboratory research techniques for compositional analysis of particles and clusters that may provide a basis for development of future instruments.

**SESSION 6. WHAT DO WE KNOW ABOUT ENGINE EXHAUST AND AIRCRAFT WAKES?**

Chairman

Dr. Michael Prather, Goddard Institute for Space Studies, National Aeronautics and Space Administration

Synopsis

A review of the CIAP studies of aircraft exhaust pointed up the difficulties in detecting the exhaust plume unless a visible trail is evident (contrail or fuel "puffs" as in YF-12 experiments). With no in situ measurements, Concorde emission studies have been limited to test-stand studies of the engines. Engine exhaust products for current engines, as well as for the new technologies that may reduce the NO₂ emission index (EI) from 40 to 5 g/kg of fuel, were described. There may be trade-offs in the new technologies between emissions of NOₓ and CO (affecting engine efficiency). Information is now available on the detailed mix of organics in engine exhaust. Wake characteristics of aircraft depended on individual aircraft dynamics.
Results were taken from wind tunnel experiments and from computational fluid dynamics work.

A number of key findings emerged from the session:

- It will be extremely difficult to measure chemical products in the exhaust plume of an aircraft without a visible clue (e.g., contrail or colored exhaust) to help find the wake. Our current database on the location of engine exhaust within aircraft wakes (the dynamically created wing-tip vortices may contain part or most of the exhaust products) is based on pictures of the visible plume (i.e., condensed water, possibly environmental) and does not necessarily describe the distribution of engine exhaust.

- It is important to re-analyze the engine test-stand results for exhaust products and to bring in new instruments or experimenters, as necessary, to address the completeness and accuracy for engine ELs of interest to HSRP/AESA.

- It is also important to characterize the size-shape distribution of soot particles from the engine tests and to compare them with the currently collected stratospheric samples. The chemical composition and surface properties of the engine soot should also be measured.

SESSION 7.  OKAY, WHAT IS OUR PLAN OF ACTION?

Chairman

Dr. Michael Prather, Goddard Institute for Space Studies, National Aeronautics and Space Administration

Questions/Issues

- Does the chemical climatology tell us where the SST exhaust goes?
- Does it tell us what the exhaust will do to the ozone?
- What instruments do we need?
- What platforms must we use?
- Is there overlap with other programs?
- Where do we need measurements?
  - Tropics?
  - Mid-latitudes?
  - Poles?
- Is the plume worth chasing?
- If so, what is our strategy?
- Do we need to be able to remote sense the plume?
Solve the Following Problems:

- Coordinate with other aircraft campaigns.
- Set priorities on instrument/platform development.

Synopsis

This final session attempted to draw conclusions from the presentations and discussion of the workshop. In particular, HSRP/AESA needs to put together a plan of action for atmospheric measurements that would be supported or encouraged by the program, i.e., WHERE DO WE GO FROM HERE?

The chair presented a sequence of key scientific questions, identified in Table 5, that were meant to focus the HSRP/AESA measurement program. These questions are listed in sequence, leading from the aircraft emissions up to the global perturbations. They are likely to be asked of the scientific community when its members are assessing the environmental impact of aircraft emissions.

The questions point to the specific measurements that are required to support the development of the assessment models and to corroborate their predictions for the present atmosphere. The discussion that followed worked on identifying the most important measurements or other studies that should be pursued in order to answer these questions.

Table 5. Key Science Questions for HSRP/AESA Assessment

<table>
<thead>
<tr>
<th>Topic</th>
<th>Question</th>
</tr>
</thead>
<tbody>
<tr>
<td>Emissions</td>
<td>What really comes out of aircraft?</td>
</tr>
<tr>
<td>Wake/Plumes</td>
<td>Can &quot;plume processing&quot; affect NET emissions?</td>
</tr>
<tr>
<td>Transport</td>
<td>How do emissions MIX and ACCUMULATE in the atmosphere?</td>
</tr>
<tr>
<td>Chemistry</td>
<td>Can we PREDICT the ozone/climate perturbations caused by additions of NOₓ, H₂O, aerosols, etc?</td>
</tr>
</tbody>
</table>

Emissions and Plume Chemistry

Before we try to measure and calibrate the emission products from engines in flight, it would be best to go back to the engine test stands from which we have derived our current values for aircraft emissions.

- For previous results, we need to check the accuracy of the reported data and the sensitivity of instruments.
For future work, we should coordinate the atmospheric experimenters with the combustion engineers and possibly make new measurements from engine test facilities with an expanded set of instruments (e.g., NO, NO₂, NO₃, aerosols, CCN, soot, OH, SO₂, SO₃, ions, HC, HCOₓ).

In-flight validation of the ground tests may be necessary as part of the political/regulatory process, but is not an immediate priority for the HSRP/AESA studies, since no new ideas have been put forward that need testing or might alter our present understanding of the wake.

It may be important to measure the chemistry and physics (aerosols) within the exhaust plume of an aircraft. Such measurements could examine chemical processing in the unique, highly concentrated environment before the exhaust plume disperses into the background stratosphere. (They also would provide in-flight validation of EIs, a lower priority, as noted previously). Some modeling sensitivity studies have shown that global perturbations are not sensitive to chemical processing within the plume (e.g., conversion of exhaust NOₓ to HNO₃ has little impact on ozone depletion), but it is possible that ice crystal growth within the core of the exhaust plume (and subsequent fallout) may lead to transport of combustion products to lower altitudes. Furthermore, plume measurements provide a "laboratory" for testing heterogeneous chemical reactions across a large range of aerosol and trace gas concentrations under stratospheric conditions.

A clear first step is to pursue the modeling studies (both global and plume) to understand better the conditions in which plume processing has a global impact.

Chemical tracer measurements in aircraft exhaust plumes are not now viewed as a high priority. If they become necessary in the future, then we need to know more about the dynamics of the aircraft wake and the distribution of exhaust products in order to plan a successful field measurement campaign. For example, do the majority of the engine exhaust products roll up into the small (~4 m) wing-tip trailing vortices, or into the larger (~100 m) visible contrail? Some of these uncertainties might be resolved with fluid dynamical modeling or with fluid experiments.

Experimental knowledge about subsonic aircraft wakes might be expanded on a theoretical basis to supersonic wakes.

Overall, field measurements may not be necessary.

If they were necessary, we would require a remote sensor (e.g., a kind of lidar) to locate the wake in real time and some very high frequency instruments for the exhaust products (CO₂, H₂O, CCN, NOₓ) as well as O₃. Perhaps the only way to quantify measurements of this type would be to put some type of chemical tracer into the fuel and then measure all of the other species relative to that tracer.

Global Transport

The critical region for assessing model transport will be in the lower stratosphere, where the projected aircraft would fly, but the exhaust products are expected to mix to some extent throughout the stratosphere. The rate of upward mixing is important, because ozone destruction from added NOₓ is more effective at higher altitudes. The rate of downward mixing is equally important, since it controls the steady-state accumulation of exhaust products.
- Measurements that are used to deduce tracer transport within the lower stratosphere must extend from the upper troposphere (about 10 km) to middle stratosphere (about 25 km), from Equator to pole, and in all seasons.

What platforms do we have or anticipate that could make these measurements? The ER-2 (full in situ instrumentation) has an operating range of about 15-20 km. The Concorde (potential for some in situ monitoring) flies regularly over the North Atlantic at 15-18 km. Lower altitudes could be filled by commercial aircraft such as the DC-8 (10 to 12.5 km), but in situ sampling is not as complete as for the ER-2. The gap between 20 and 25 km will technically be filled by UARS and its correlative measurements, but the range of vertical overlap (redundancy) with the extensive ER-2 data will be small, and the need for in situ measurements for some species and for some tests is not met.

- There is a clear need for additional platforms (either balloons or a Perseus/HAARP-type aircraft) to make measurements above ER-2 flight levels. The need to reach greater altitudes is important, but the currently anticipated Perseus/balloons have limited payloads (i.e., only a few species of the ER-2 in situ suite can be measured concurrently).

- The reconstruction and interpretation of aircraft campaign data will require concurrent measurements (or analyzed data) of

1. dynamical tracers (pressure, temperature, potential temperature, potential vorticity),

2. ozone and water vapor, as well as

3. one or more long-lived tracers (N2O, CFCs, CO2, CH4).

The comparison of these observations with model simulations (in order to test the model's predictions for aircraft emissions) will require corresponding predictions of chemical tracer transport variations from 3-D models.

The impact of aircraft flying in the stratosphere today (the Concorde corridor over the North Atlantic at flight levels 500-600; commercial fleets at flight levels 310-390 over the poles in winter) may be detectable. Because of the altitude restrictions on both the ER-2 and the DC-8, some of the interesting altitudes are not presently reachable. The asymmetries in stratospheric NOy H2O climatologies between the Arctic and Antarctic may be related to the denitrification/desiccation over Antarctica or may represent aircraft input.

- A full climatology of CO2-H2O-NOy and O3 in the mid-latitude stratosphere might be able to discern a factor (appropriate enhancements of CO2-H2O-NOy) that matches aircraft exhaust.

It would be extremely useful to develop a unique measure of aircraft perturbation (e.g., Is stratospheric soot a unique product of aircraft, and can it be measured with adequate frequency and accuracy?).

The following examples of trace species or types of measurements were discussed as being important in diagnosing stratospheric transport:

- Soot and CCN spectrum as a measure of current engines

- Is there an isotopic signature (e.g., 13C/12C, D/H) in Jet-A fuel?
Transport through tropopause folds, cyclonic events, polar vortex
Gradients and removal processes in the upper troposphere
Natural cycles of H₂O and NOₓ
Stratospheric NOₓ from tropospheric lightning

Chemical Perturbations

We will need to measure the important chemically active species under as many possible conditions in today's atmosphere in order to test our model predictions of stratospheric photochemistry under highly perturbed conditions predicted for an HSCT fleet (i.e., high NOₓ, H₂O, CO, soot, etc.). One important issue would be to verify the crossover region where the addition of NOₓ leads to NOₓ-catalyzed loss of ozone above about 15 km and to smog-chemistry production of ozone below about 10 km (see Figure 1).

- Along with measurements of the source gases (noted above for determining the global transport and dispersion of aircraft exhaust), we will need to measure the NO-NO₂-HNO₃-NOₓ family, the OH-HO₂ concentrations, and the ClO-BrO-CINO₂ family.

If we achieve the extent of coverage cited above for transport, then we will likely have sampled a sufficiently large range of photochemical environments.

Uncertainty in the kinetics and radiation data for the models could be reduced by atmospheric measurements of specific chemical balances (e.g., the NO₂-NO₃-N₂O₅ equilibrium, the HOₓ-NOₓ chemistry, of course, along with other key tracers such as O₃, H₂O, CH₄, NOₓ, Clₓ). Likewise, certain atmospheric conditions such as mountain lee waves provide an accessible test of heterogeneous chemical processing. It is not clear whether these atmospheric tests would provide clues to missing chemistry. A most difficult question is, how do we determine the role of added aerosols both locally (along aircraft corridors), at the winter poles (in PSCs), and at or near the tropical tropopause?

The following examples of other trace species or types of measurements would be important in diagnosing stratospheric chemistry and its perturbations:

- Look for effects of CCN on PSCs as well as cirrus/radiation
- Determine the natural stratospheric sulfur cycle and particle formation
- Determine molecular clusters that might affect J-values
- Measure UV-visible scattered light to test model photolysis
APPENDIX

LOWER STRATOSPHERIC MEASUREMENT ISSUES:
A WORKSHOP
NASA Ames Research Center
October 17-19, 1990

Agenda

MAJOR QUESTIONS:
What atmospheric measurements are needed for an SST assessment?
Can we make those measurements?

Wednesday, October 17

Welcome
Logistics
Overall High Speed Research Program (HSRP) Perspective
Define Atmospheric Effects of Stratospheric Aircraft Program

A. Schmeltekopf/NOAA Retired
E. Condon/NASA Ames
H. Wesoky/NASA HQ
M. Prather/NASA GISS

Known Problems in Lower Stratospheric Chemistry

Chairman’s Introduction
Homogeneous Reactions
Heterogeneous Reactions
Global Chemical Modeling
Plume Modeling
Panel Discussion

C. Howard/NOAA AL
S. Sander/NASA JPL
M. Tolbert/SRI International
S. Wofsy/Harvard
C. Kolb/Aerodyne

W. Brune/Penn State

Questions:
What critical laboratory measurements are needed?
What critical atmospheric measurements are needed?
Can in-situ measurements help us directly determine rates?

Known Problems in Lower Stratospheric Transport

Chairman’s Introduction
Large-Scale Dynamics
Stratosphere-Troposphere Exchange
Radiative Forcing
Global Transport Modeling
Plume Mixing
Tracer Database: What Can We Learn?
Panel Discussion

J. Holton/U. Washington
J. Holton/U. Washington
M. Schoeberl/NASA Goddard
D. Crisp/Caltech
A. Plumb/MIT
M. Prather/NASA GISS
H. Johnston/U. C. Berkeley
E. Danielsen/NASA Retired
A. Tuck/NOAA AL

Additional Panel Members:

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Questions:
Will SST exhaust really mix upward?
Can the high-resolution structure of the transport be important?
Can exotic tracer experiments be useful?

What Platforms Do We Have or Can We Get?

Chairman's Introduction
ER-2
Condor
Perseus
HAARP
Concorde as a Platform

Panel Discussion (Platforms)
Additional Panel Members:

Questions:
Do any of these platforms get us far enough?
High enough?
Long enough (duration)?
When can the platforms be ready?

Thursday, October 18

Present Measurement Capabilities

Chairman's Introduction
Radicals
Source Gases
Aerosols
Dynamic Tracers
Radiation
Panel Discussion
Additional Panel Members:

Questions:
Are all of our present capabilities accurate enough?
Specific enough?
Fast enough?
Do we currently have the "Right Stuff?"

New Measurement Techniques

Chairman's Introduction
Lidar
Microwaves
Aerosols
Diode Laser Spectrometer

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Aerosols
CO₂
OH/HO₂
Two-Step Laser Mass Spectrometry for
Analysis of Adsorbates and Particulates
Ion Spectroscopy and Dynamics
Spectroscopy and Reactivity of Clusters
Panel Discussion

Questions:
Which of these new areas look promising enough to warrant development for
use in atmospheric research?
Can they be ready by 1992?
1994?
When?

Friday, October 19

What Do We Know about Engine Exhaust?

Chairman's Introduction
Wake Studies in CIAP
Concorde
Wake Chemicals
Wake Characteristics

Okay, What Is Our Plan of Action?

Chairman's Introduction
Panel Discussion
Additional Panel Members:

Questions:
Does the chemical climatology tell us where the SST exhaust goes?
Does it tell us what the exhaust will do to the ozone?
What instruments do we need?
What platforms must we use?
Is there overlap with other programs?
Where do we need measurements?
Tropics?
Mid-latitudes?
Poles?
Is the plume worth chasing?
If so, what is our strategy?
What instruments do we need?
Do we need to be able to remote sense the plume?

Solve the following problems:
Define 1992 and 1994 missions
Coordinate with other aircraft campaigns
Set priorities on instrument/platform development