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Trace Gas Study Accumulates Forty Million Frequent-Flyer Miles for Science

At any given moment about 4000 commercial aircraft are flying over the Earth. To an atmospheric scientist they constitute a potentially vast array of in situ measurement platforms, an obvious source of high-resolution upper tropospheric/lower stratospheric (UT/LS) trace gas data that could not be obtained on such a continuous basis in any other way, especially over the oceans.

The number of flights has been steadily growing, and the long-term outlook for continued "funding" looks excellent given the penchant for humans to travel around the world. And yet, to date, there have been few attempts to tap this resource for atmospheric chemistry studies. One study, however, has chalked up 40 million miles over the past 5 years.

Using commercial airliners to measure trace gases in the atmosphere is a well-justified, self-monitoring activity, since engine exhaust is constantly injecting material into the flight corridors. Although the total amount is not great (2% to 3% of all anthropogenic atmospheric pollution), the UT/LS region is sensitive to aircraft emission in several ways. Nitrogen oxides, CO, and hydrocarbons through photochemical reactions may increase the amount of tropospheric ozone, which is a greenhouse gas; and water vapor, soot, and sulfur oxides can increase contrail clouds and aerosols that may modify UT/LS radiative forcing and the global temperature.

With air traffic expected to double in the next 10 to 15 years, aviation effects on the atmosphere can only be expected to increase as well. To take advantage of this existing atmospheric sampling infrastructure, a program to measure ozone and water vapor by Airbus in-service aircraft (MOZAIC) was launched in January 1993 [*Marenco et al.*, 1998]. So far MOZAIC has recorded over 10,300 flights totaling 74,000 hours and 40 million miles. Initial scientific results were published in a special section of the *Journal of Geophysical Research* in 1998 (vol. 103, no. D19) as well as in other journals, including two papers in *Nature*.

First Project in 1968

The first project to use in-service aircraft for trace constituent data collection took place in 1968 with carbon monoxide observations made from Lufthansa B-707 airliners [*Seiler and Junge*, 1970]. From the vertical CO gradient observed above the tropopause, the researchers estimated the size of the stratospheric CO sink. (The same group employed a manned laboratory inside a container on 10 German Cargo Services flights between 1981 and 1987 to measure CO.)

Also, under the tropospheric ozone (TROZ) campaign, the meridional distribution of ozone was studied using 37 commercial flights between northern Europe and South Africa during 1970-1974. That study concluded that upper tropospheric ozone distribution was determined primarily by input from the stratosphere [*Fabian and Pruchniewicz*, 1977].

NASA's global atmospheric sampling program (GASP) was the first continuous, long-term effort to piggyback trace gas measuring instruments on commercial jetliners. Four B-747s (two Pan American, one United, and one Qantas) were equipped with instruments to routinely measure ozone, carbon monoxide, water vapor, aerosols, temperature, and horizontal winds [see, e.g., *Nastrom*, 1979].

The program lasted from 1975 to 1979 with over 6900 flights (80% between 30°N and 55°N) recorded. Highlights included estimates of the vertical and horizontal flux of ozone near the tropopause, interhemispheric transport deduced from CO results, the discovery of upper tropospheric low-ozone regions caused by Walker circulation transport from the western Pacific boundary layer, the refutation of the mesoscale kinetic energy gap, and demonstration of terrain effects on flight-level mesoscale wind variability.

Now, after nearly 2 decades, commercial airliners are again being used for collecting trace gas data. Carbon monoxide, carbon dioxide, and methane were measured routinely between April 1993 and July 1996 on a Japan Airlines B-747 flying between Japan and Australia [*Matsueda et al.*, 1998]. Twelve air samples were taken during each flight and analyzed later in a laboratory. Results showed that the seasonal cycles of these trace gases differ significantly between the Northern and Southern Hemispheres and that southern African or South American biomass burning signatures appear in the upper troposphere over Australia in the spring.

Another study, the nitrogen oxides and ozone measurements along air routes (NOXAR) investigation, was conducted from May 1995 to May 1996. Instruments were operated on 540 Swissair B-747 flights in the Northern Hemisphere, with data recorded every 3 s for a horizontal resolution of about 750 m at cruise altitude. One of the initial major findings of NOXAR was the detection of large-scale (100-1300 km) nitrogen oxide plumes in the upper troposphere that were produced by continental pollution or lightning [*Brunner et al.*, 1998]. The elevated levels of ozone within the plumes, particularly in the summer, suggested an enhancement in photochemical ozone production because of the high nitrogen oxide concentration.

Although GASP generated a formidable amount of data on the UT/LS region, data qual-

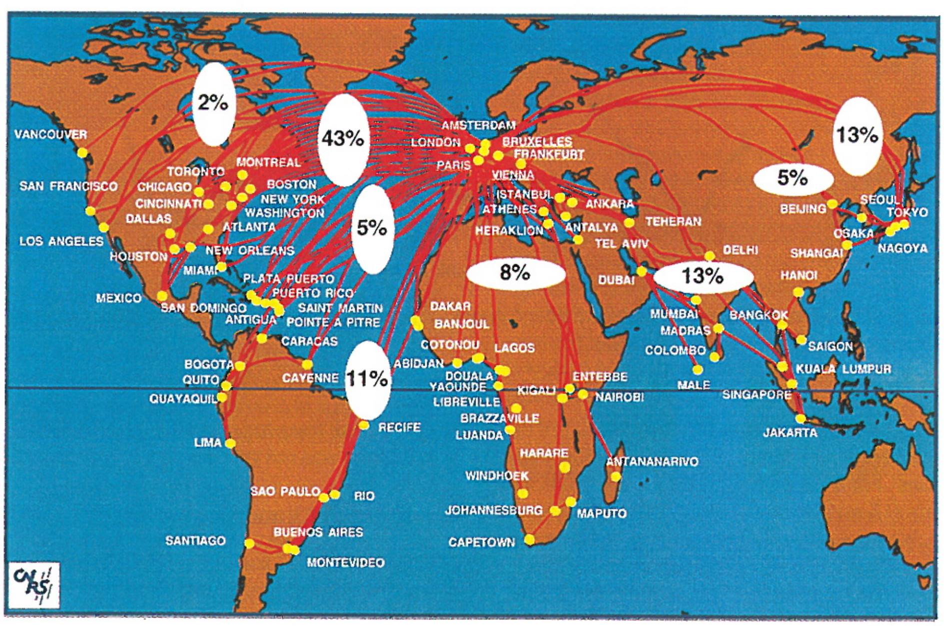

Fig. 1. Coverage map of MOZAIC flights. Schematic flight routes are shown as red lines. The percentages of flights to and from the various global sectors are also displayed.

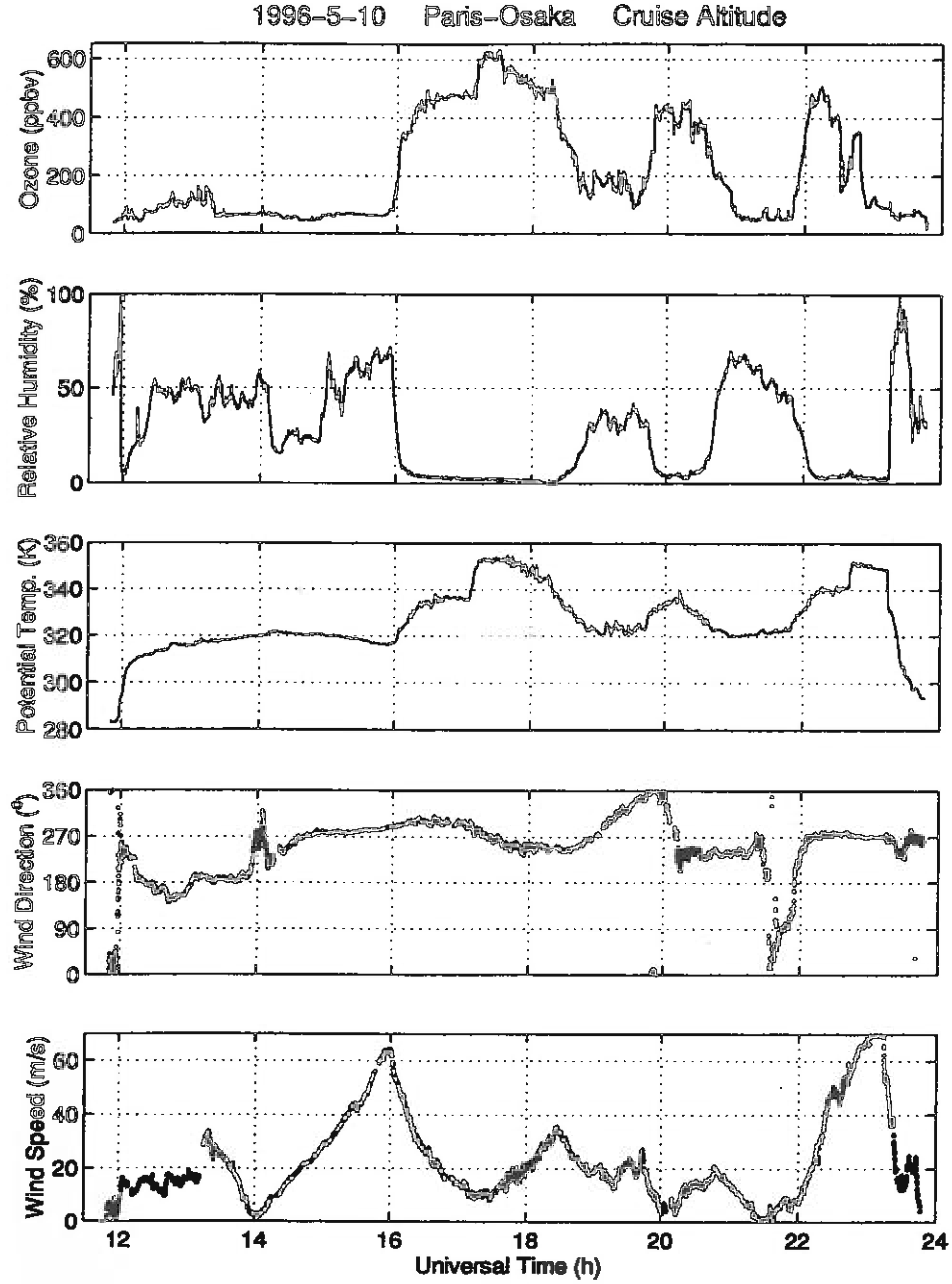

Fig. 2. Time series of ozone, relative humidity, potential temperature, and horizontal winds from a Paris-to-Osaka flight at cruise altitude.

ity and resolution issues hindered their full use for scientific purposes, especially for the trace gases. For most flights the data were collected only at 5-min intervals, there were instrumental problems with the CO instrument due to water condensation, and there were uncertainties about the water vapor data quality. Moreover, these experiments were performed during a time when theories on photochemical formation of ozone were just developing and their significance was not yet universally recognized.

Quasiglobal Climatology

Sensing the need for a new GASP-like program that utilized updated technology, European scientists in collaboration with Airbus Industrie (and its partners), Air France, Austrian Airlines, Lufthansa, and Sabena initiated the MOZAIC program in January 1993. They hoped to establish a quasiglobal climatology of the large-scale distributions of ozone and water vapor in the UT/LS region and to determine the vertical and temporal distributions of ozone and water vapor at various airport locations around the world. They also wanted to investigate the seasonal and geographical variations of the measured distributions in relation to their natural and anthropogenic sources, stratosphere-troposphere exchanges, and horizontal and vertical air circulations. The scientists hoped as well to validate and improve the current three-dimensional (3D) chemistry and transport models used to evaluate the present and future impact of human activity by comparison between MOZAIC climatologies and model outputs, with particular emphasis on subsonic aircraft emissions.

MOZAIC's modus operandi was to keep the instrument package small and light to minimize the cost load for the carriers. For the first phase, ozone, water vapor, and temperature probes were integrated into a rack that was roughly equivalent to the space and weight of one passenger plus baggage (full weight 117 kg). The package was installed in the avionic compartment below the cockpit, with the intake tubes mounted on the fuselage 7 m back from the aircraft's nose. The system operates automatically without help from the flight crew. However, the airline maintenance crew does check for malfunctions during routine stopovers. Time, position, pressure, airplane speed, and horizontal wind data are obtained from the aircraft's air data computer. All the data are recorded at 4-s intervals from takeoff to landing.

The processed and validated data are stored in a database at the Centre National de Recherche Météorologique, Toulouse, operated by Météo-France. To aid in data interpretation, daily maps of meteorological and satellite data are also archived. Potential vorticity and tropopause characteristics interpolated every minute along each aircraft trajectory also are available. Booklets containing data plots from each flight and daily meteorological fields are available free of charge to MOZAIC coinvestigators (see end of article on how to become a coinvestigator).

The global distribution of the MOZAIC flight routes is shown in Figure 1. Clearly the Atlantic Ocean sector is well covered, but even the 8% for Africa represents over 820 flights, a vast improvement in coverage over TROZ or GASP for that region. Figure 2 shows example data from a flight at cruise altitude. On this flight, the aircraft clearly went through stratospheric air during three intervals when ozone and potential temperature were very high and humid-

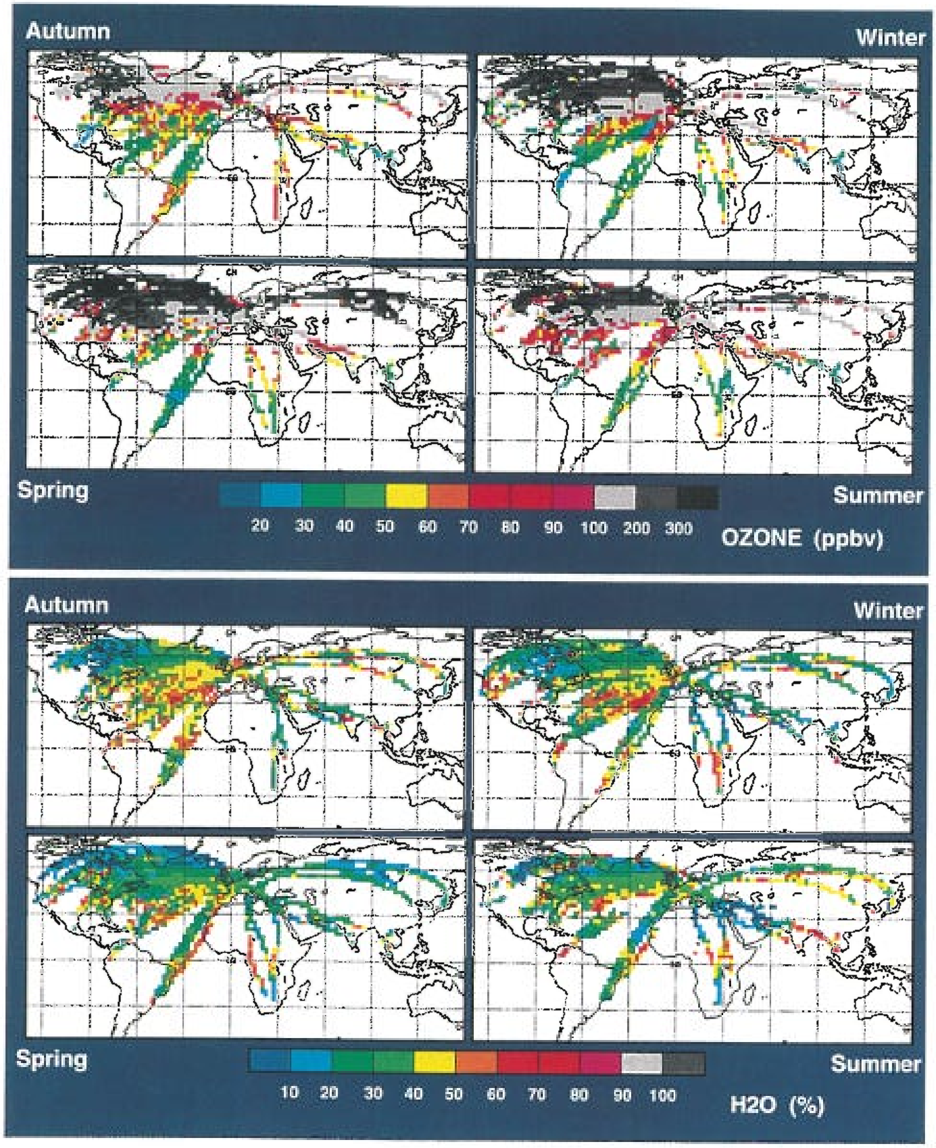

Fig. 3. Geographical and seasonal distributions in ozone concentration (top) and relative humidity (bottom) at the 238-hPa pressure flight level (10.6 km) from MOZAIC data collected between September 1994 and August 1996 (autumn, S-O-N; winter, D-J-F; spring, M-A-M; summer, J-J-A). The horizontal resolution of the plotted data is 2.5°.

ity was extremely low. Also the horizontal winds changed markedly at times when the troposphere-stratosphere transitions took place.

Initial Results Successful

MOZAIC has certainly been successful in establishing a climatology of ozone in the UT/LS region over a wide range of latitudes and longitudes (see Figure 3; relative humidity climatology is also displayed). This climatology was established for five flight levels between 9 and 12 km altitude. To distinguish between stratospheric (ozone-rich) and tropospheric (ozone-poor) air, a threshold of 100 ppbv of ozone was used in a first attempt. An analysis of the seasonal variations in both the UT and LS regions was made possible in this way, and the results highlighted the spring maximum/winter minimum in stratospheric air and the summer maximum/fall minimum in tropospheric air.

At northern mid-latitudes, the distribution of ozone was related to the intensities of dynamic (tropopause variations, stratosphere/troposphere exchanges, variations of the polar front, and the position of ridge/trough pressure systems) and photochemical processes. At tropical latitudes, the MOZAIC climatology showed the influences of active photochemical processes, deep convection, and biomass burning emissions [*Thouret et al.*, 1998].

This climatology is important for understanding dynamic and chemical processes involved in the upper tropospheric ozone distribution, for 3D model validations (only MOZAIC data are available at these altitudes with such wide spatial coverage), for comparisons with other ozone measurements (ozone soundings, other airborne programs, satellite data), and for estimation of flight times within the stratosphere. The horizontal wind and temperature fluctuation data can also be used to study gravity waves and 2D turbulence and the atmospheric kinetic energy spectrum in general. The comparison between the 2-year MOZAIC ozone climatology (1994-1996) and the long series of some stations from the ozone sounding network has shown that they were in rather good agreement, despite the different natures of the programs. Mean concentrations derived from ozonesonde series are about 3% to 13% higher that those obtained by MOZAIC in the free troposphere; these differences are within the range of uncertainties of the two techniques.

Recent modeling studies have demonstrated that aircraft contrails have the potential for climate forcing. However, a quantitative assessment has been difficult because contrail parameterization has been problematic. The MOZAIC humidity and temperature fluctuation data have helped in this regard. The fluctuation distributions together with local criteria for the formation and persistence of contrails have allowed calculation of the maximum fractional coverage of contrails within a model grid [*Gierens et al.*, 1997].

The study also showed that large-scale distributions of relative humidity and temperature fluctuations could not be modeled as Gaussians. Instead, they manifested long tails that were closer to Lorentzian in form.

Ozone measurements during cross-equatorial MOZAIC flights have confirmed the dynamical barrier effect of the subtropical tropopause to exchanges between the subtropical stratosphere and the tropical troposphere. However, it seems that the barrier could be leaky at small scales (<100 km) as ozone transients are observed south of the subtropical jet in the upper equatorial troposphere [*Suhre et al.*, 1997].

Dramatic Expansion of Coverage

Because every flight includes a takeoff and a landing, vertical profiles of the measured quantities are produced over the departure and arrival airports; an example is shown in Figure 4. Tropospheric layers of ozone and other trace gases have been observed and analyzed using data from research aircraft missions, but

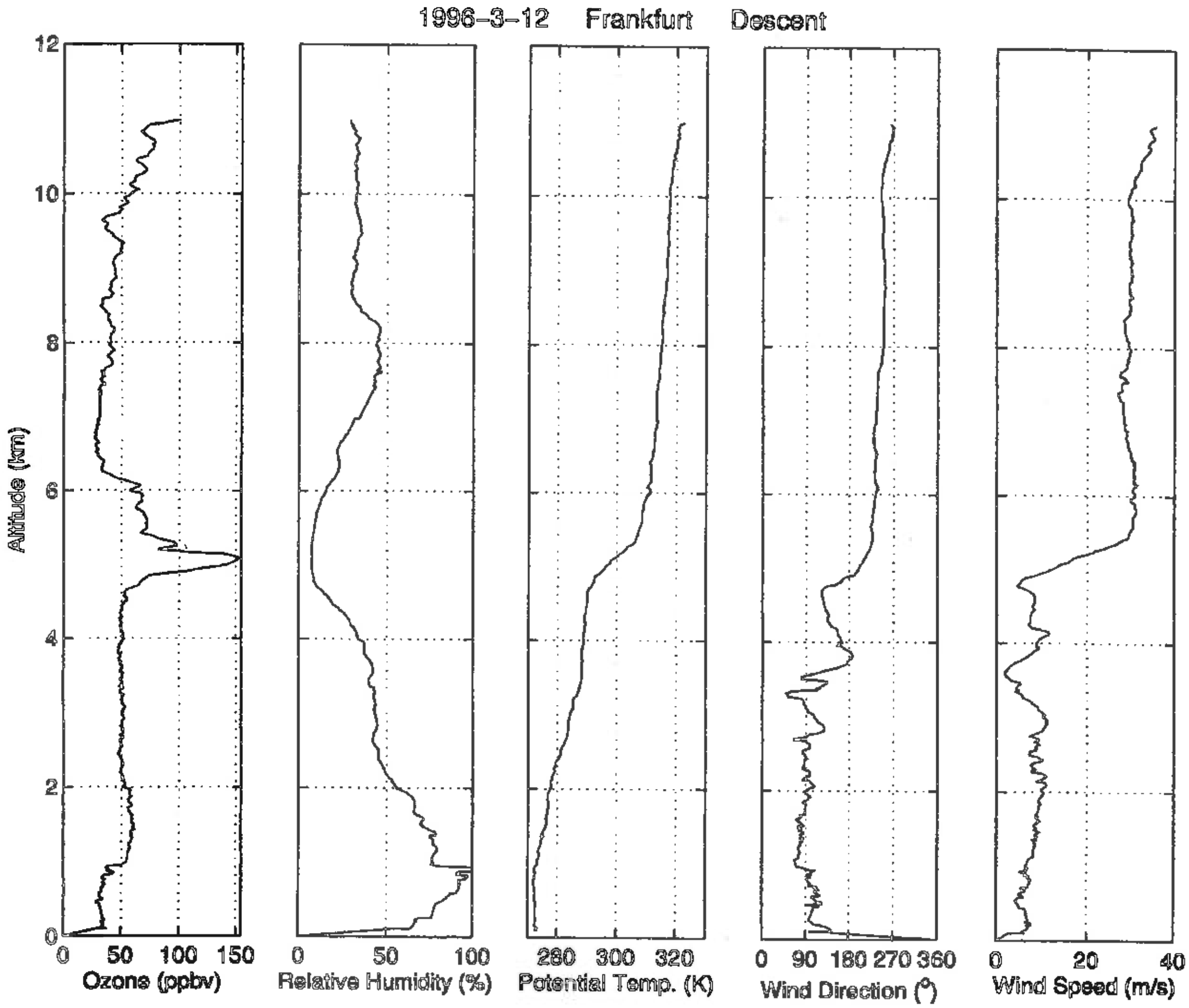

Fig. 4. Vertical profiles of ozone, relative humidity, potential temperature, and horizontal winds obtained during descent into Frankfurt, Germany. Note the high-ozone, low-humidity layer at 5 km, which was also associated with enhanced static stability and wind shear. Most likely it originated in the stratosphere.

val airports; an example is shown in Figure 4. Tropospheric layers of ozone and other trace gases have been observed and analyzed using data from research aircraft missions, but MOZAIC has dramatically expanded the global and seasonal coverage. Layers can be categorized according to the various combinations of high or low anomalies in the trace gas levels with respect to the background concentrations. For example, with two constituents, ozone and water vapor, there would be four types: low ozone/low humidity, low ozone/high humidity, high ozone/low humidity, and high ozone/high humidity.

Newell et al. [1999] found from MOZAIC data that approximately half of all layers everywhere (including the tropics) were of the high ozone/low humidity type (Figure 4). Most likely this type of layer originates in the stratosphere (since the stratosphere has much higher ozone levels and is much dryer than the troposphere) and thus has implications for stratosphere-troposphere exchange. However, to really confirm the source of this and other types of layers, one needs to measure other marker species such as CO and examine the 3D dynamical context in which the layer was formed and advected. This kind of detailed study is being performed using dedicated research aircraft (NASA's ongoing Pacific Exploratory Mission (PEM); see Newell et al. [1999] for references).

The high quality and fine time resolution of the MOZAIC ozone data allow the study of small-scale features. For example, an important characteristic of the layers is that they are sometimes as thin as a few hundred meters. Chemical reaction rates can be highly nonlinear, so the output of an averaged input quantity is not necessarily equal to the averaged output of multiple inputs at finer resolution. Such resolution dependency has shown up quite strongly in model calculations of chemical ozone loss. Current models are unable to resolve these thin layers, which places a fundamental limit on their accuracy. Thin humidity layers can also have local radiative forcing effects. Dry layers are self-stabilizing; wet layers are destabilizing. Patches of clear-air turbulence may be generated by such unstable layers.

New Phased Proposed

MOZAIC has gone through two phases and, to extend the program several years more, a third phase has been proposed to the Fifth Framework Program of the European Communities for 2000-2002. MOZAIC-III plans to add CO, NO_y, aerosols, vertical wind velocity, and turbulence measurements.

These new measurements will considerably enhance the possibilities of interpretations in the UT/LS. Possible will be evaluation of convective transport of pollutants from the continental boundary layer into the upper troposphere; climatology of nitrogen species; discrimination between stratospheric and tropospheric air (combined with O_3, H_2O, and potential vorticity criteria); better identification of tropospheric layer characteristics based on criteria similar to those used for PEM data; better validation of 3D chemical and transport models; and large-scale distributions of aerosols and turbulent structure of the atmosphere. The extension of O_3 and water vapor time series also will offer the possibility to study trends and interannual variabilities in the UT/LS.

Besides meeting its own programmatic scientific goals, it is MOZAIC's philosophy to maximize the scientific use of its database. A huge reservoir of high-quality observations, the database is indeed of valuable interest for the international scientific community. Powerful tools have been developed to allow easy access and fast recovery of the data by authorized users via ftp and the Web.

Interested scientists are encouraged to become coinvestigators in the program by developing collaborations with MOZAIC participants under rather flexible conditions—submission of a scientific project and signing of the MOZAIC Data Protocol. Information on the program and on the procedure to follow are available on the Web (http://www.aero.obs-mip.fr/mozaic/) and from Alain Marenco, Laboratoire d'Aérologie, Observatoire Midi-Pyrénées, 14 Avenue Edouard Belin, 31400 Toulouse, France; E-mail: mara@aero.obs-mip.fr.

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