The Brewer–Dobson circulation in the light of high altitude in situ aircraft observations


1NOAA Aeronomy Laboratory, USA
2National Center for Atmospheric Research, USA
3NASA Ames Research Center, USA
4NOAA Climate Monitoring and Diagnostics Laboratory, USA
5CIRES, University of Colorado, USA
6JPL/Caltech, USA
7University of Wisconsin, USA
8University of Denver, USA

(Received 18 March 1996; revised 23 May 1996)

(Symons Memorial Lecture: delivered by A. F. Tuck on 17 May 1995)

SUMMARY

Fast response in situ measurements of a suite of chemical species made from the NASA ER2 high altitude aircraft, between 60°N and 70°S at potential temperatures up to 530 K from March to November 1994 at longitudes 115°W to 150°E, are considered for the view they offer of the Brewer–Dobson circulation in the lower stratosphere and upper troposphere. In the southern hemisphere, where most of the flights occurred, comparisons are made with measurements taken in August/September 1987 at longitudes 120°W to 60°W to examine temporal and longitudinal differences. Interpretations made suggest conceptual modifications to the simple construct of advection in a two-dimensional long-term mean.

KEYWORDS: Airborne observations Atmospheric chemistry Stratospheric circulation Troposphere–stratosphere interaction

1. INTRODUCTION

It is 50 years to the month since the completion of the series of frost-point hygrometer profiles of water vapour which formed the basis upon which the Brewer–Dobson circulation was proposed (Dobson et al. 1946; Brewer 1949), as Professor Brewer reminded me in the tea interval. Those flights, performed by Boeing Flying Fortress and de Havilland Mosquito aircraft, showed that the air above the tropopause over southern England could not have been dried by ascent through the local tropopause. In a remarkable piece of detective work, it was proposed that stratospheric air was dried by ascent through the tropical tropopause, with possibilities such as gravitational separation of gases being eliminated by the observation of stratospheric carbon dioxide (CO₂) at ~ 300 parts per million by volume (p.p.m.v.) via a modified frost-point instrument; the argument that the profiles over

* Corresponding author: United States Department of Commerce, NOAA Aeronomy Laboratory, Environmental Research Laboratories, 325 Broadway, Boulder, Colorado 80303-3328, USA.
southern England were representative was buttressed by back-trajectory analysis, showing that, on a time-scale of a few days, the air had originated over Canada and not the tropics.

While the Brewer–Dobson circulation, with rising air at low latitudes followed by poleward motion and descent, has stood the test of time as a gross long-term, zonal–mean construct, it triggered a long debate over the exact mechanisms for stratospheric dehydration, occasioned by the observation that the lower stratosphere was drier (< 6 p.p.m.v.) than the saturation mixing ratio over ice at the temperature and pressure of the zonal–mean tropical tropopause (≈ 20 p.p.m.v., 200 K, 90 hPa). While water vapour has proven to be relatively easy to detect in the context of the range of chemical species currently being examined, it has also proved to be extraordinarily difficult to measure accurately, given that some arguments hinge on differences of a few tenths of a p.p.m.v., or about 10% of the average stratospheric mixing ratio. The elimination of contamination by outgassing and of temperature gradients within instruments proved to be important; by the end of the 1960s, propelled by the need to understand the fate of radioactive debris from atmospheric nuclear-weapon tests, there was a general agreement that the lower stratosphere was dry (< 6 p.p.m.v. between 30 and 100 hPa). The transport of water vapour, ozone and radioactive debris could be qualitatively understood by early zonal–mean calculations of the stratospheric circulation, in a Lagrangian sense by Murgatroyd and Singleton (1961) and in an Eulerian sense by Reed and German (1965). Observational studies of the correlations between radioactivity, ozone and potential vorticity (PV) led to the notion that downward transfer of air from the stratosphere occurred in mid latitudes by tropopause folds produced during upper-tropospheric cyclogenesis in association with jet streams (Reed and Danielsen 1957); Danielsen (1968) pointed out that the values of ozone in and above such folds implied prior quasi-isentropic transport from a region of strong diabatic descent, namely the winter polar vortex. Brewer and Wilson (1968) showed that if a calculation of the stratospheric ozone content was performed with known insolation and absorption cross-sections for a pure oxygen atmosphere (diluted 1:4 by N₂), an excess of ozone by a factor of 2 or 3 was predicted. The total ozone column moreover would be incorrectly distributed, with a maximum in the tropics and minima at the poles (Dütsch 1971). These results highlighted, respectively, the need for extra sinks and the importance of the circulation.

With the advent of concerns about the photochemical stability of the stratospheric ozone abundance under perturbations of water vapour (Harrison 1970) and nitrogen oxides (Johnston 1971) from the jet engines of proposed supersonic civil airliners, there was a great increase in stratospheric research activity. This was boosted still further by the idea that chlorine atoms and chlorine monoxide produced by photodissociation of chlorofluorocarbons (CFCs) (Molina and Rowland 1974) could propagate an ozone-destroying chain reaction even more efficient than those proposed earlier for OH and HO₂ from water (Hampson 1965) and for NO and NO₂ from aircraft (Johnston 1971) and nitrous oxide (Hampson 1964, 1966; Crutzen 1970, 1971). The key notion of excited oxygen atoms reacting with H₂O and N₂O to produce OH, HO₂, NO and NO₂ which could then propagate an ozone-destroying chain reaction in the stratosphere appears to have been put forward in Hampson’s articles, following the laboratory discovery of the role of O(¹D) in the flash photolysis of water and ozone by McGrath and Norrish (1960). The work up to the early 1980s has been reviewed in a number of places (CIAP 1975; COMESA 1975; Harries 1976; Ellsaesser 1979; World Meteorological Organization 1986). Two discoveries of note were the realization that the stratosphere was a source of water vapour, via the oxidation of methane (Wofsy et al. 1972), and of the hygropause, a region of minimum water situated a few kilometers above the tropical tropopause in Brazil and later found to be present in mid latitudes (Kley et al. 1979). The need to understand water vapour quantitatively, and
to provide insight into the mechanisms of entry to and exit from the stratosphere, caused
Danielsen et al. (1982) to propose equipping a modified Lockheed U2R aircraft (known to
the National Aeronautics and Space Administration (NASA) as the ER2) with fast response
in situ instruments to measure temperature, pressure, winds, total water and water vapour,
ozone, reactive nitrogen, carbon monoxide, condensation nuclei (CN) and ice particles.
The proposal was funded by NASA, and the aircraft was used to study transport around jet
streams over North America in 1984 (Danielsen et al. 1991) and processes connected with
tropical cumulonimbus clouds north of Australia in early 1987 (Danielsen 1993). At short
notice, the ER2 was equipped with a new instrument to measure chlorine monoxide (ClO)
(Brune et al. 1988), and with the CO instrument modified to measure N₂O (nitrous oxide)
instead, this payload was deployed to investigate the ‘ozone hole’ (Farman et al. 1985)
from Punta Arenas in the Straits of Magellan during August and September 1987, the
results of which have been reported in the Airborne Antarctic Ozone Experiment (AAOE)
This mission was the first airborne examination of the winter vortex with in situ instru-
mentation, and it produced some important results and controversial ideas. The loss of
ozone was shown to be unequivocally caused by chlorine (Anderson et al. 1989a; Jones
et al. 1989), the partitioning of which between the reactive (Cl and ClO) and inactive
(HCl, ClONO₂) forms of chlorine had been shifted by exposure to the surfaces of polar
stratospheric clouds (PSCs) (Solomon et al. 1986), observations of the NO₃ content of
which were shown to be consistent (Fahey et al. 1989) with a theoretical prediction of the
behaviour of mixed nitric acid–water particles which formed at temperatures 5–8 K above
frost-point (Toon et al. 1986). The vortex was also shown to contain large masses of air in
which both water vapour and total reactive nitrogen had been substantially depleted, pre-
sumably by gravitational sedimentation of crystals containing H₂O and HNO₃ and which
were large enough (∋ 4 μm) to fall out of the stratosphere, or at least to altitudes below
about 350 K potential temperature (Kelly et al. 1989; Fahey et al. 1990). The dehydrated
and denitriﬁed vortex was also shown to contain very low values of tracers such as CFCs,
N₂O and CH₄, indicating substantial diabatic descent (Heidt et al. 1989; Loewenstein et
al. 1989). The vertical structure of the tracers was shown to have a transition at potential
temperature θ ∼ 400 K (Tuck 1989; Podolske et al. 1989). A controversial notion which
arose during the interpretation of the observations was that the vortex was approximated
better as a ﬂow reactor rather than as a containment vessel. On the basis of the overall
observational database, and citing an analysis by Murphy et al. (1989) as evidence for both
horizontal exchange at the edge of the vortex and diabatic descent within it, Tuck (1989)
proposed that there was signiﬁcant mass ﬂow through the vortex, both horizontally in the
range of 475 > θ > 400 K and vertically through the θ ≈ 400 K level (the tropopause was
at ≈ 300 K). The unexpectedly dry air between about 40°S and the vortex edge (located on
average at about 65°S) was also invoked as evidence for the spread of vortex air (Kelly et
al. 1989). This idea had been examined earlier by Elsaaesser (1975), using results taken by
a WB57F aircraft in October 1972. Proffitt et al. (1989a, b) deﬁned the vortex edge, and
decided, largely on the basis of N₂O–O₃ correlations, that there was substantial mass ﬂow
through the outer vortex, mainly via inﬂow of lower-latitude air above and outﬂow via
diabatic descent to below θ ∼ 400 K, in which region there was freer exchange with mid
latitudes than was the case above. The containment-vessel view, originally proposed from
theoretical modelling by Juckes and McIntyre (1987), was supported by Schoeberl et al.
(1992) and implicitly assumed in a chemical kinetic analysis of ozone loss (Anderson et
al. 1989b). This view essentially relies on the argument that low rates of horizontal mixing
are balanced by low diabatic cooling rates (dθ/dt ∼ 0.5 K day⁻¹) to yield a near-ﬁxed
mass of air in the vortex, while the ﬂow reactor has larger mixing rates balanced by larger
cooling rates \((d\theta/dt > 1.0 \text{ K day}^{-1})\). Danielsen and Houben (1988) had concluded that there must be substantial descent in the vortex, and predicted radial gradients of tracers such as CFC-11 and CH\(_4\).

The observation during the AAOE that the outer part of the antarctic vortex had temperatures similar to those characteristic of the interior of the arctic vortex, and showed signs of PSC-processed air and ozone loss (Tuck 1989; Proffitt et al. 1989c), caused a similar mission (the Airborne Arctic Stratospheric Experiment (AASE)) to be deployed from Stavanger, Norway during January and February 1989. The arctic vortex indeed had high levels of active chlorine (Brune et al. 1990) and in that year some patchy denitrification (Fahey et al. 1990; Kawa et al. 1992), and so was reported to exhibit ozone loss in the 10\%-35\% range at ER2 altitudes (Proffitt et al. 1990; McKenna et al. 1990; Salawitch et al. 1990). The issue of containment versus flow reactor remained controversial, with Kelly et al. (1990) reporting a water vapour asymmetry between the northern and southern wintertime hemispheres and interpreting it as evidence for flow through the vortices; Tuck et al. (1992) showed evidence for the peel-off of vortex filaments into northern mid latitudes in both the aircraft data and PV maps from operational meteorological analyses, the latter of which were too coarse to match the high horizontal resolution of the observations. On the other hand, Schoeberl et al. (1992) argued the case for containment in the southern vortex.

There was agreement that after these two missions there was a need to examine the vortices over their whole life cycle, not just at the peak period for chemical ozone loss. After some debate, the arctic was investigated first during the boreal winter of 1991–92 (AASE-II; Anderson and Toon 1993) and the antarctic during the austral winter of 1994 by the airborne Southern Hemisphere Ozone Experiment/Measurements for Assessing the Effects of Stratospheric Aircraft (ASHOE/MAESA; NASA 1994). The objectives of these two missions were expanded by the laboratory discovery that the normal sulphuric-acid aerosol in the Junge layer could activate chlorine, via an indirect temperature-independent process converting N\(_2\)O\(_5\) to nitric acid (Tolbert et al. 1988) and also by some direct temperature-dependent processes which convert HCl and ClONO\(_2\) to forms which readily photolyze to Cl and ClO (Hanson and Ravishankara 1991, 1994). The large volcanic eruption of Mount Pinatubo in June 1991 occurred a few weeks before the start of the AASE-II, which thus served to investigate the effects of the large amounts of sulphuric-acid aerosol produced from the SO\(_2\) injected by the volcanic plume. An extra mission, the Stratospheric Photochemistry, Aerosol and Dynamics Experiment (SPADE), was performed during November 1992 and April/May 1993, with two new instruments measuring CO\(_2\) and OH/HO\(_2\) (Wofsy et al. 1994; Boering et al. 1994; Wennberg et al. 1994). Other important developments were the realization that the PSCs seen during the AASE in January 1989 were not explicable by the formation of nitric acid trihydrate crystals (Dye et al. 1992), and the theoretical development of high-resolution representations of the dynamics of PV on an isentropic surface, with the filamentation being characterized as chaotic advection (Pierce and Fairlie 1993; Waugh and Plumb 1994; Norton 1994; Sutton et al. 1994; Waugh et al. 1994), the defining property of which is the exponential stretching of material lines of fluid.

In recent years there has been a revival of interest in the possibility of flying supersonic airliners in the stratosphere; one important question raised by such a possibility is the extent to which the emissions from such aircraft could be transported from the primary place of injection, namely mid latitudes, into the tropics where they could rise towards the main ozone-production region. There is thus an interest in the transport of air between the tropics and mid latitudes as well as between the poles and mid latitudes, and advantage was taken of the transit flights between California and New Zealand to investigate this with flights from Hawaii and Fiji. This lecture will be mainly concerned with presenting an overview
of the results from the ASHOE/MAESA, appealing where necessary to results from the earlier AAOE, AASE, AASE-II, and SPADE missions. The ASHOE/MAESA data will be analysed more thoroughly in many papers related to individual instruments or subsets of instruments over the next few years.

2. AIRCRAFT PAYLOAD AND FLIGHT PLANS

The payload used on most of the ASHOE/MAESA flights is shown in Fig. 1; there were several flights where the HO$_3$, NO$_2$, ACATS (Airborne Chromatograph for Atmospheric Trace Species) and ClO/BrO instruments were removed, and the HIS (High-resolution Interferometric Sounder) fitted instead. The former configuration will be referred to as the full payload, the latter as the dynamics and radiation payload. The instruments whose observations are used in this paper are listed in Table 1. A list of ER2 flights during the airborne ozone missions since 1987 is given in Table 2, and a catalogue of the flights comprising the ASHOE/MAESA is given in Table 3. It should be noted that not all instruments were intended to function fully on the test flights, and that the flights of 940320, 940415 and 940608 were curtailed at an early stage because of aircraft problems (dates are given in yymmd format, hence 20 March, 15 April and 8 June 1994). The ASHOE/MAESA data cover latitudes from 60°N to 70°S, and 15 to 21 km (360 < θ < 540 K) with profiles to the surface for some instruments above the airfields. All horizontal data were taken above ~18 km in the ASHOE/MAESA, and above 16 km during the AAOE and the AASE.

From the main base in Christchurch during 1994 at ER2 cruise levels, θ > 440 K, the flight-level temperature decreased in both poleward and equatorward directions over the 26° radius of action. Thus normal aircraft cruise climb outbound was approximately isentropic, with a few exceptions when very cold air was present at the south end in late July and early August. The return leg was generally performed with the aircraft climbing as fast as possible so as to intersect the maximum number of isentropes. At the maximum

![Figure 1](image-url). The NASA ER2 high altitude research aircraft and its ‘full’ payload for the ASHOE/MAESA. For the dynamics and radiation flights the ClO, HO$_3$, NO/NO$_2$ and ACATS instruments were removed, and the HIS instrument (see Table 1) was added in a belly pod. See text for further explanation.
TABLE 1. INSTRUMENTAL CHARACTERISTICS

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Technique</th>
<th>Error Limits</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>H2O</td>
<td>Lyman-α/fluorescence</td>
<td>6%</td>
<td>Kelly et al. (1989)</td>
</tr>
<tr>
<td>O3</td>
<td>UV absorption</td>
<td>3%</td>
<td>Proffitt et al. (1989a)</td>
</tr>
<tr>
<td>NO2</td>
<td>Chemiluminescence</td>
<td>&lt; 20%</td>
<td>Fahey et al. (1989)</td>
</tr>
<tr>
<td>N2O</td>
<td>IR tunable diode laser</td>
<td>10%</td>
<td>Loewenstein et al. (1989)</td>
</tr>
<tr>
<td>CH4; HCl; CO</td>
<td>IR tunable diode laser</td>
<td>6-10%; 0.1-0.2 p.p.b.v.; 6-10%</td>
<td>Webster et al. (1994)</td>
</tr>
<tr>
<td>CO2</td>
<td>Non-dispersive IR</td>
<td>0.1 p.p.m.v.</td>
<td>Boering et al. (1994)</td>
</tr>
<tr>
<td>CFCl3; CH4; H2</td>
<td>EC gas chromatograph</td>
<td>3%+precision</td>
<td>Elkins et al. (1996)</td>
</tr>
<tr>
<td>Condensation nuclei</td>
<td>Butanol/laser</td>
<td>15%</td>
<td>Wilson et al. (1983)</td>
</tr>
<tr>
<td>u, v, T, p</td>
<td>Probes/INS</td>
<td>1 m s⁻¹; 0.3 K; 0.3 kPa</td>
<td>Scott et al. (1990)</td>
</tr>
<tr>
<td>T(z)</td>
<td>57-59 GHz O2</td>
<td>1 K to ±1.5 km; 3 K to ±5 km</td>
<td>Denning et al. (1989)</td>
</tr>
<tr>
<td>Irradiance †</td>
<td>FTIR 3.8-16.6 μm</td>
<td>1 K in retrieved T</td>
<td>Smith (1991)</td>
</tr>
</tbody>
</table>

TABLE 2. SOURCES OF AIRBORNE OBSERVATIONS

<table>
<thead>
<tr>
<th>Mission</th>
<th>Date</th>
<th>Airfield</th>
<th>No. of take-offs</th>
</tr>
</thead>
<tbody>
<tr>
<td>AAOE</td>
<td>Aug.–Sep. '87</td>
<td>Moffett NAS (37°N, 122°W)</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Howard AFB (9°N, 80°W)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Puerto Montt (41°S, 73°W)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Punta Arenas (53°S, 71°W)</td>
<td>13</td>
</tr>
<tr>
<td>AASE</td>
<td>Dec. '88–Feb. ‘89</td>
<td>Moffett NAS (37°N, 122°W)</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Wallops Island (38°N, 75°W)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Stavanger (59°N, 6°E)</td>
<td>15</td>
</tr>
<tr>
<td>AASE-II</td>
<td>Oct. '91–Mar. ‘92</td>
<td>Moffett NAS (37°N, 122°W)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Elson AFB (65°N, 105°W)</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Bangor (44°N, 69°W)</td>
<td>19</td>
</tr>
<tr>
<td>ASHOE/MAESA</td>
<td>Feb.–Nov. ‘94</td>
<td>Moffett NAS (37°N, 122°W)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Barber’s Point NAS (21°N, 158°W)</td>
<td>7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Nadi (18°S, 177°E)</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Christchurch (44°S, 173°E)</td>
<td>27</td>
</tr>
</tbody>
</table>

distance from the airfield, or at an intermediate distance on some transit flights, a profile was performed with descent at 1000 ft min⁻¹ to 50 000 ft, followed by ascent at the same rate back to cruise level, a procedure referred to as a ‘dip’. Its purpose was to provide vertical information, within the limits of safe aircraft operation. Forecast maps of PV were electronically available for flight planning in real time at Christchurch, as were high horizontal resolution contour-advection forecasts on selected isentropes. The forecasts were crucial for aligning the aircraft track to optimize both vortex penetration and the chance of encountering filaments of air peeled off from both vortex and tropics. They were performed by teams from NASA Langley, NASA Goddard, UK Meteorological Office, UK Universities Global Atmospheric Modelling Project, University of Wisconsin and the Australian Climate Research Centre/Massachusetts Institute of Technology, using analyses from the UK Meteorological Office (UKMO), European Centre for Medium-Range Weather Forecasts (ECMWF), NOAA* National Meteorological Center (NMC), NASA Goddard Space Flight Center and the Australian Weather Bureau.

3. THE UPWARD BRANCH: TROPICS

(a) Horizontal structure

An early observation during the ASHOE/MAESA was that there is an ‘inner tropics’, characterized by near-tropospheric mixing ratios of molecules possessing a source at the surface and a sink in the middle to upper stratosphere, such as N2O, CH4 and CFCs. The presence of sharp horizontal gradients in NO2, O3 and their ratio at the edges of the tropics

* National Oceanic and Atmospheric Administration.
<table>
<thead>
<tr>
<th>Date</th>
<th>Description</th>
<th>From</th>
<th>To</th>
<th>Approx. duration (hours)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>940121</td>
<td>TestFIt MASP/FCAS</td>
<td>Ames</td>
<td>Ames</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940202</td>
<td>D&amp;R TestFIt</td>
<td>Ames</td>
<td>Ames</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940204</td>
<td>D&amp;R TestFIt-&gt; 59°N</td>
<td>Ames</td>
<td>Ames</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940214</td>
<td>TestFIt</td>
<td>Ames</td>
<td>Ames</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940218</td>
<td>TestFIt-&gt; 55°N</td>
<td>Ames</td>
<td>Ames</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940219</td>
<td>TestFIt</td>
<td>Ames</td>
<td>Ames</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940314</td>
<td>TestFIt MMS only</td>
<td>Ames</td>
<td>Ames</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940318</td>
<td>TransitFIt</td>
<td>Ames</td>
<td>B Pt</td>
<td>5+</td>
<td>no dip</td>
</tr>
<tr>
<td>940320</td>
<td>FltSth-&gt; 16°N</td>
<td>B Pt</td>
<td>B Pt</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940321</td>
<td>FltSth-&gt; 2°S</td>
<td>B Pt</td>
<td>B Pt</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940322</td>
<td>FltSth-&gt; 6°N</td>
<td>B Pt</td>
<td>B Pt</td>
<td>5</td>
<td>2 dips</td>
</tr>
<tr>
<td>940327</td>
<td>TransitFIt</td>
<td>B Pt</td>
<td>Nadi</td>
<td>7</td>
<td>no dip</td>
</tr>
<tr>
<td>940329</td>
<td>TransitFIt</td>
<td>Nadi</td>
<td>Chch</td>
<td>4+</td>
<td>T &lt; ~86 °C; 1 dip</td>
</tr>
<tr>
<td>940330</td>
<td>FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940403</td>
<td>FltNth-&gt; 19°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940405</td>
<td>FltSth-&gt; 65°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940408</td>
<td>FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940413</td>
<td>D&amp;R FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940415</td>
<td>D&amp;R FltNth-&gt; 38°S</td>
<td>Chch</td>
<td>Chch</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940523</td>
<td>FltSth-&gt; 47°S</td>
<td>Chch</td>
<td>Chch</td>
<td>2+</td>
<td></td>
</tr>
<tr>
<td>940524</td>
<td>FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940528</td>
<td>FltNth-&gt; 21°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>940601</td>
<td>FltSth-&gt; 65°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>bottomdip</td>
</tr>
<tr>
<td>940603</td>
<td>FltSth-&gt; 67°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>ClO elimbout</td>
</tr>
<tr>
<td>940605</td>
<td>D&amp;R FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>no HIS</td>
</tr>
<tr>
<td>940608</td>
<td>D&amp;R FltSth-&gt; 45°S</td>
<td>Chch</td>
<td>Chch</td>
<td>1</td>
<td>max alt 47 kft</td>
</tr>
<tr>
<td>940728</td>
<td>FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>PSC</td>
</tr>
<tr>
<td>940730</td>
<td>FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>PSC</td>
</tr>
<tr>
<td>940801</td>
<td>FltNth-&gt; 19°S</td>
<td>Chch</td>
<td>Chch</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>940806</td>
<td>FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>O3 less 67°S</td>
</tr>
<tr>
<td>940808</td>
<td>D&amp;R FltSth-&gt; 69°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>dehydr dip</td>
</tr>
<tr>
<td>940810</td>
<td>D&amp;R FltSth-&gt; 67°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>941003</td>
<td>D&amp;R FltSth-&gt; 68°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>vortex edge 53°S</td>
</tr>
<tr>
<td>941005</td>
<td>D&amp;R FltSth-&gt; 65°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>941008</td>
<td>FltEvonNZ</td>
<td>Chch</td>
<td>Chch</td>
<td>4</td>
<td>Concorde wake</td>
</tr>
<tr>
<td>941010</td>
<td>FltSth-&gt; 70°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>941013</td>
<td>FltSth-&gt; 69°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td>HCl = Cl2 in dip</td>
</tr>
<tr>
<td>941016</td>
<td>FltSth-&gt; 70°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>941020</td>
<td>FltSth-&gt; 64°S</td>
<td>Chch</td>
<td>Chch</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>941022</td>
<td>TransitFIt</td>
<td>Chch</td>
<td>Nadi</td>
<td>7+</td>
<td>dip at 10°S</td>
</tr>
<tr>
<td>941024</td>
<td>TransitFIt</td>
<td>Nadi</td>
<td>B Pt</td>
<td>7</td>
<td>dip at 3°S</td>
</tr>
<tr>
<td>941026</td>
<td>FltSth-&gt; 2°N</td>
<td>B Pt</td>
<td>B Pt</td>
<td>7</td>
<td>H2O layers in dip</td>
</tr>
<tr>
<td>941029</td>
<td>FltSth-&gt; 2°S</td>
<td>B Pt</td>
<td>B Pt</td>
<td>8</td>
<td>H2O layers in dip</td>
</tr>
<tr>
<td>941102</td>
<td>TransitFIt</td>
<td>B Pt</td>
<td>Ames</td>
<td>6+</td>
<td>1 dip; headwind</td>
</tr>
<tr>
<td>941104</td>
<td>FltNth-&gt; 59°N</td>
<td>Ames</td>
<td>Ames</td>
<td>8</td>
<td>HCl/Cl2 &gt; in SH</td>
</tr>
</tbody>
</table>

B Pt = Barber's Point Naval Air Station
Chch = Christchurch International Airport
Ames = Moffett Field Naval Air Station

was reported by Proffitt et al. (1989c) and Murphy et al. (1993); here we show in Figs. 2(a) and (b) the horizontal traces of N2O and NOy (which is produced in the course of N2O loss) during the flight of 940327 from Hawaii (21°N, 158°W) to Fiji (17°S, 177°E). There are sharp gradients at about 7°N and 12°S with a lesser change at 8°S which separate air from the mid-latitude stratosphere at the potential temperatures of the flight track, from air of more recent tropospheric origin near the equator. Note that the inner tropics in this ‘snapshot’ are only 37% of the width of the geographically defined tropics. Further, note also that at the flight level of θ ≈ 470 K, the value of N2O is significantly less than that characteristic of the troposphere in 1994 (≈ 300 p.p.b.v. (parts per billion (10⁶) by volume) vs. 311 p.p.b.v.), a point to which we shall return in the ensuing section on vertical structure. Figure 3 shows the PV structure for the trans-tropics flight track on 940327. Because of
Figure 2. Horizontal traces during the flight of 940327 (potential temperature $\geq 440$ K) from Hawaii to Fiji of (a) N$_2$O and (b) NO$_x$. (c) NO$_y$ vs. N$_2$O scatterplot.

Figure 3. Potential vorticity (PV) and potential temperature ($\theta$) along the Hawaii–Fiji flight track of 940327. $\theta$, solid lines in 25 K increments from 350 to 675 K; PV is dashed for negative values, solid for positive. The aircraft climbed from $\theta \approx 460$ K near Hawaii to 480 K near Fiji in acquiring the observations shown in Fig. 2.
the change of sign in planetary vorticity at the equator the isopleths of PV start to bow upwards in the inner tropics; the co-location of this region with the inner tropics as defined by the tracers can be seen by comparing Figs. 2 and 3. Note that in both cases the region of most recent tropospheric tracers and vertical PV isopleths is displaced towards the vernal hemisphere. Figures 4 and 5 show the trans-tropical flight of 941024 and the PV along the track; comparison with Figs. 2 and 3 shows the shift between March and October. Note, however, that there is also a higher fraction in the geographical tropics (i.e. equatorward of 23°37' latitude) populated by mid-latitude air, presumably as the result of weaker wave activity in the recently past winter southern hemisphere balanced by weaker tropical convection during that period.

Figure 2(c) shows the N₂O–NOₓ scatterplot for 940327. There are two distinct branches at values of N₂O less than 295 p.p.b.v.; the one with relatively high NOₓ values is composed mainly of points measured north of latitude 7°N, while that with relatively low NOₓ is of points measured mainly south of latitude 5°S. There are populations of points which connect these two populations, indicating some mixing. Many of these points, plus virtually all the points with 295 < N₂O < 311 occur in the inner tropics, where the anti-correlation is less tight. There is a population of points with N₂O ≈ 298 p.p.b.v. and NOₓ ≈ 1 p.p.b.v. at 16°S at θ ≈ 450 ± 10 K, an indication of the transport of air from the inner tropical zone. It is not immediately clear why the northern tropics should contain

![Figure 4](image_url)

Figure 4. Horizontal traces during the flight of 941024 (potential temperature ≈ 440 K) from Fiji to Hawaii of (a) N₂O and (b) NOₓ. (c) NOₓ vs. N₂O scatterplot.
Figure 5. Potential vorticity (PV) and potential temperature ($\theta$) along the Fiji–Hawaii flight track of 941024. Contour labelling is as in Fig. 3. In obtaining the data in Fig. 4 the aircraft climbed from $\theta \approx 460$ K near Fiji to $\theta \approx 480$ K near Hawaii, with a dip to $\theta \approx 350$ K near the equator.

Figure 6. Mean horizontal structure of $N_2O$ from 60°N to 70°S during (a) phase 1 and (b) phase 4 of the ASHOE (potential temperature $\geq 440$ K). The averages in this and subsequent figures are composed of the data taken on the flight dates below the figures. Bars are standard deviations.
more NO$_3$ relative to N$_2$O than the southern tropics. We will return to this subject later. This asymmetry was not observed during October (Fig. 4(c)), when the entire curve was characteristic of the northern branch (i.e. high NO$_3$) in March.

Figure 6 shows the mean horizontal structure of N$_2$O as a function of latitude, averaging together the flights shown by date in the figure legend separately for the phase 1 (mid February/late March) and phase 4 (late October/early November) observations.

The structure between 15 and 40° latitude at both times of year suggests significant movement into this belt from both the inner tropics and the polar regions. We show here the mean horizontal structure in three other quantities; CO$_2$ in Fig. 7, 2CH$_4$ + H$_2$O in Fig. 8 with CH$_4$ and H$_2$O separately in Fig. 9. CO$_2$ has a similar gross structure to N$_2$O (Fig. 6), but for a different reason: it has no sinks in the stratosphere, but a large enough increasing trend in the troposphere to yield a mean decrease with height above the tropopause, whereas N$_2$O has a similar stratospheric morphology arising from the presence of photochemical sinks in the middle and upper stratosphere, particularly at lower latitudes. The structure of 2CH$_4$ + H$_2$O is similar to CO$_2$ and N$_2$O, a fact which raises an interesting question. The morphology of methane (Fig. 9) in the lower stratosphere is very similar to that of nitrous oxide, for essentially the same reasons. Water vapour (Fig. 9(b)) is somewhat lower and more variable in the tropics than in middle and subpolar latitudes, an observation consistent with the notion of tropical drying at the tropopause. The sum 2CH$_4$ + H$_2$O should, however, be conservative in the absence of trends in either water vapour or methane (Jones et al. 1986) or of significant stratospheric production of molecular hydrogen, H$_2$. The measurement of

![Figure 7](image_url)
Figure 8. Same as Fig. 6, but for $2\text{CH}_4 + \text{H}_2\text{O}$. The CH$_4$ data are from ALIAS.

H$_2$ during phase 4 eliminates this latter possibility: its mixing ratio is nearly constant at $\sim 0.5$ p.p.m.v., and the latitudinal structure of $2\text{CH}_4 + \text{H}_2\text{O} + \text{H}_2$ (Fig. 10) is essentially identical to that of $2\text{CH}_4 + \text{H}_2\text{O}$ in Fig. 8(b). The tropospheric trend in methane is far too small to account for the lower values outside the inner tropics, in the air which is ‘older’ by an average of 3–4 years (Volk et al. 1996). The obvious explanation is that there has been an increase in the amount of water vapour entering the stratosphere between about 1990 and 1994, amounting to 0.5 p.p.m.v. Essentially the same result is obtained by using ALIAS* CH$_4$ rather than those for ACATS (Fig. 11) and it is evident at $440 < \theta < 480$ K at the end of both northern winter (March 1994) and northern summer (October 1994). The result agrees in sign with that of Oltmans and Hofmann (1995) from mid-latitude time series of frost-point hygrometer ascents. It is interesting to note that the period 1990–95 was coincident with the longest El Niño on record.

(b) Vertical structure

We now turn to the vertical structure in trace species in the tropical region, and portray it by considering the mean vertical profiles of N$_2$O above Hawaii, near the equator, and above Fiji. Figure 12(a) shows the three profiles for March, while Fig. 12(b) shows them in late October/early November. In all cases there is a decrease with height from the lowest potential temperatures (340–360 K) to the highest (450–480 K), with the slopes above about 400 K being steeper at the edges of the tropics (Hawaii and Fiji) than near

* Airborne Laser Infrared Absorption Spectrometer.
Figure 9. ASHOE phase 4 (potential temperature \( \geq 440 \) K) averages from 60°N to 70°S. (a) ALIAS CH\(_4\) and (b) H\(_2\)O. These separate traces were combined to produce Fig. 8(b).

Figure 10. ASHOE phase 4 (potential temperature \( \geq 440 \) K) averages from 60°N to 70°S. The quantity plotted is 2CH\(_4\) + H\(_2\)O + H\(_2\), where the methane and hydrogen measurements were obtained by ACATS.

the equator. The steepest such slope is above Hawaii at the end of northern winter. The context in which to view the absolute mixing ratios in Fig. 12 is provided in Table 4, which gives the monthly mean surface values of CO\(_2\), N\(_2\)O and CFC-11 at Mauna Loa (20°N, 156°W) and American Samoa (14°S, 171°W) during 1994, as measured by the NOAA Climate Monitoring and Diagnostics Laboratory network. The mean values everywhere on the N\(_2\)O profiles, which range from the upper tropical troposphere to the lower tropical
stratosphere, are less than the values at the surface, even those in the southern hemisphere. These results seem to indicate that in this range of potential temperatures, there is some import of older, $N_2O$ depleted air from the mid-latitude stratosphere into the upper tropical troposphere (recall that the lifetime of $N_2O$ against photodissociation at 20 km near the equator is many years, 63 according to Volk et al. (1996)). The curves in Fig. 12 are undoubtedly entirely stratospheric at the top end, and tropospheric at the very bottom. In between, however, the profiles may average together upper tropospheric and lower stratospheric air, owing to the flight-to-flight variation of the potential temperature of the tropopause. Accordingly, the data are replotted in Fig. 13 using pressure altitude relative to the tropopause in place of $\theta$. Note that the tropopause value is always less than the lowest surface value, and there is always a decrease with height in the upper troposphere. These upper-tropospheric slopes are shown for $N_2O$ and CFC-11 in Table 5. The implication
TABLE 4. Monthly mean surface mixing ratios of CO₂, N₂O, CFC-11 from Mauna Loa Observatory (MLO, 20°N, 156°W) and American Samoa (SMO, 14°S, 171°W) during 1994

<table>
<thead>
<tr>
<th>Month</th>
<th>CO₂ (p.p.m.v.)</th>
<th>N₂O (p.p.b.v.)</th>
<th>CFC-11 (p.p.t.v.)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MLO</td>
<td>SMO</td>
<td>MLO</td>
</tr>
<tr>
<td>01</td>
<td>357.84</td>
<td>356.73</td>
<td>310.9</td>
</tr>
<tr>
<td>02</td>
<td>358.46</td>
<td>356.97</td>
<td>310.9</td>
</tr>
<tr>
<td>03</td>
<td>359.28</td>
<td>356.91</td>
<td>310.9</td>
</tr>
<tr>
<td>04</td>
<td>360.59</td>
<td>356.70</td>
<td>311.0</td>
</tr>
<tr>
<td>05</td>
<td>361.29</td>
<td>356.77</td>
<td>311.1</td>
</tr>
<tr>
<td>06</td>
<td>360.30</td>
<td>356.94</td>
<td>311.3</td>
</tr>
<tr>
<td>07</td>
<td>358.56</td>
<td>356.89</td>
<td>311.3</td>
</tr>
<tr>
<td>08</td>
<td>356.58</td>
<td>356.82</td>
<td>311.4</td>
</tr>
<tr>
<td>09</td>
<td>354.93</td>
<td>356.93</td>
<td>311.0</td>
</tr>
<tr>
<td>10</td>
<td>355.11</td>
<td>357.25</td>
<td>311.1</td>
</tr>
<tr>
<td>11</td>
<td>357.00</td>
<td>357.70</td>
<td>311.2</td>
</tr>
<tr>
<td>12</td>
<td>359.10</td>
<td>358.11</td>
<td>311.3</td>
</tr>
</tbody>
</table>

Figure 13. Same data as in Fig. 12, but replotted using pressure altitude relative to the tropopause as the vertical coordinate.

of Fig. 13 and Table 5 is that there is recirculation of aged stratospheric air from middle latitudes into both the upper troposphere and the lower stratosphere. The greatest extent and variability of such recirculation is seen above the tropopause in March, particularly in the profiles of N₂O and CO₂, consistent with the notion of the greatest effect being as a result of the largest dynamical eddy activity during northern winter. The effect is more clearly evident over Hawaii and Fiji, both of which are equatorward of the mean latitude of the subtropical jet stream (27°), than near the equator. Figure 14(a) shows the March profiles of N₂O, CO₂ and CFC-11 relative to the tropopause. The CFC-11 profile looks different to the others for two reasons: there are no CFC-11 data for 940322, and because CFC-11 has a shorter lifetime in the stratosphere than the others, and so has a steeper decrease with height. Figure 14(b) shows the equatorial profiles for October, at the end of the southern winter. Above the tropopause, both N₂O and CFC-11 show evidence of
### TABLE 5. Mean upper-tropospheric slopes and mean tropopause values of CO₂, N₂O, CFC-11 near Hawaii, the equator (±7°), and Fiji during late March and late October 1994

<table>
<thead>
<tr>
<th></th>
<th>CO₂ (p.p.m.v.)</th>
<th>N₂O (p.p.b.v.)</th>
<th>CFC-11 (p.p.t.v.)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Hawaii</td>
<td>Equator</td>
<td>Fiji</td>
</tr>
<tr>
<td>Slope below tropopause per km March</td>
<td>-0.44</td>
<td>-0.15</td>
<td>-0.48</td>
</tr>
<tr>
<td>Mixing ratio at tropopause</td>
<td>356.73</td>
<td>356.83</td>
<td>356.57</td>
</tr>
<tr>
<td>Slope below tropopause per km October</td>
<td>0.07</td>
<td>ns</td>
<td>-0.01</td>
</tr>
<tr>
<td>Mixing ratio at tropopause</td>
<td>356.33</td>
<td>356.77</td>
<td>357.20</td>
</tr>
</tbody>
</table>

ns=no significant slope obtainable.
nd=no data.

---

![Figure 14](image-url)

Figure 14. Mean vertical profiles of N₂O, CO₂ and CFC-11 relative to the tropopause near the equator (0° ± 7° latitude): (a) phase 1 and (b) phase 4 of the ASHOE. The flights contributing to the averages are shown with the figures. Note that the values are all less than the surface values in Table 4.

Horizontal transport from mid latitudes, although to a lesser extent and with less variability than in March. The structure in the CO₂ has been interpreted by Boering et al. (1994) as the propagation of the seasonal variation in the northern hemisphere troposphere up to 1.5–3.0 km above the tropopause during the previous 7 months. Note that the values there, 358.0 ± 0.22 p.p.m.v., are higher than any actually measured in March from the ER2, although they do correspond to the monthly mean values at Mauna Loa Observatory, Hawaii in early 1994. However, note that the CFC-11 and N₂O profiles both indicate substantial
dilution by mid-latitude air between 1500 and 3000 m above the equatorial tropopause (Fig. 14(b)) and it is difficult to reconcile this with the high CO₂ there. The N₂O and CFC-11 profiles both show positive deviations from the average slope between 2500 and 3000 m above the tropopause, and it is possible that this air may contain some fraction of more recent injection by overshooting thunderstorm tops. One possibility, that of stratospheric CO₂ production by methane loss, falls short of being able to explain 358 p.p.m.v. of CO₂ 3 km above the equatorial tropopause in air having ≈ 3% N₂O loss and ≈ 8% CFC-11 loss relative to tropospheric values. Consideration of Table 4 shows that the mean values at Mauna Loa during 1994 peaked in May at about 361 p.p.m.v. It would be possible to reduce 361 to 358 by dilution with mid-latitude air entering the tropics, a process which must happen because while the lifetimes of N₂O and CFC-11 at 16–20 km in the tropics are years, they decrease with height immediately above the tropical tropopause (Figs. 12–14). If we consider the mean profiles of the three species in March and October above Moffett Field (37°N, 122°W) and Christchurch (44°S, 173°E) (Fig. 15), we can make approximate estimates of the amount of mid-latitude air which must have been mixed into the tropics to account for the decreases with height above the equatorial tropopause. The values obtained from the N₂O and CFC-11 profiles are in the range 25%–60%. This amount, when applied to the ≈ 355 p.p.m.v. above Moffett Field and Christchurch at the potential temperatures of the CO₂ mixing ratios of 358 p.p.m.v. in the tropics, implies an injection of 359 p.p.m.v. CO₂ at some time before October. This injection moreover must have happened somewhere other than near the date-line, since the mixing ratios observed at the tropical tropopause there in March were less than 357 p.p.m.v. (Table 5). Surface values of 359 p.p.m.v. or greater were observed in the northern hemisphere during the first half of 1994 (Table 4). During this time, the Inter Tropical Convergence Zone (ITCZ) is south of the equator on average, and the CO₂ mixing ratios at American Samoa were about 356.8 p.p.m.v. If air with 358 p.p.m.v. of CO₂ in October 2–3 km above the tropopause was 75% air injected during the first 6 months of 1994 and 25% air mixed in from mid latitudes, the flow associated with the ITCZ during northern winter must inject mainly northern hemisphere air into the stratosphere, despite its mean position being well south of the equator. It may be that it is a combination of this process with some injection from overshooting thunderstorms, a possibility which was discussed earlier.

Figure 16 shows vertical profiles of H₂O, N₂O and NO₃ for 941026, very close to the equator. While an element of the maximum in the water at θ = 415 K could be a memory of the northern summer’s warmer tropopause, it is also apparent that some of it arises from the transport into the tropics of older mid-latitude air, as evidenced by the lower N₂O and higher NO₃ between 410 and 430 K, relative to the profiles above and below. It is clear that recirculation of air between mid latitudes and the tropics vitiates to some extent attempts to interpret the circulation by advection in a meridional plane, particularly in regard to defining the ‘age’ of stratospheric air as an accurate quantity.

The species, measured on the ER2, which shows the largest gradients between the northern and southern hemispheres is carbon monoxide, CO. It is clear from Fig. 17 that the values at the inner tropical tropopause are much less than those characteristic of the lower troposphere of either the northern hemisphere (∼ 200 p.p.b.v.) or the southern hemisphere (∼ 50 p.p.b.v.). CO has a lifetime of about 2 months in the troposphere; the main sink is reaction with OH, which will be more effective in summer than in winter, and in the lower tropical troposphere. In March the tropopause values at 12°S–25°S and 12°N–12°S are 25–30 p.p.b.v., and 35–40 p.p.b.v. in October. These tropical tropopause values are accompanied by decreases with height for 4 km or more in the upper tropical troposphere, showing that recirculation of stratospheric air is an important process. During October, the CO vertical profiles show a maximum 2 km below the tropopause in the southern subtropics
and above Christchurch, suggesting a return flow associated with monsoon circulations from north to south across the equator. Note, however, that the mixing ratios are still much less than those characteristic of the northern hemisphere lower troposphere. Finally, it is interesting to note that the mid-latitude CO mixing ratio is greater in the northern hemisphere than in the southern hemisphere, even up to 10 km above the tropopause, i.e. in the stratosphere. The stratospheric difference was more pronounced during March than
Figure 16. Vertical profiles of H$_2$O, N$_2$O and NO$_x$ for 941026, as a function of potential temperature, taken near the equator. (Bottom dip—end dip).

Figure 17. Mean vertical profiles of CO, plotted as pressure altitude relative to the tropopause, for 37°N, 12°N–25°N, 12°N–12°S, 12°S–25°S and 44°S during (a) phase 1 and (b) phase 4 of the ASHOE. (c) Mean CO from 60°N–70°S for phase 1 and phase 4 at potential temperature $\theta > 440$ K. The peak at 20°S in March was above typhoon ‘Usha’.
October. The effect may reflect quasi-isentropic transport into the stratosphere across the subtropical jet-stream latitudes, a process which would result in more CO in the northern hemisphere stratosphere. This is evident from Fig. 17(c), which shows horizontal traces of CO from 60°N to 70°S for phases 1 and 4, respectively, at the ends of the boreal and austral winters. Irrespective of season, there is more CO in the northern hemisphere lower stratosphere than in the southern. The implication appears to be that the stratosphere in each hemisphere is fed predominantly from the troposphere in its own hemisphere, presumably by a mixture of recirculated stratospheric air and air recently lofted from the lower troposphere by deep convection, entering through the inner tropical tropopause. The persistence of the asymmetry to the highest latitudes sampled indicates that some fraction of tropical air at these levels is mixed out over the entire hemisphere.

We can examine the extent of dilution of inner tropical air by horizontal transport from mid latitudes by an independent method: the use of air-parcel trajectories calculated from global meteorological analyses. Figure 18 shows the results of calculating 880 three-dimensional back trajectories from the ECMWF analyses for the 2-month periods before 940330 and 941031, initiated at each standard pressure level from 10 to 300 hPa. The curves show the percentages of air in the inner tropics, defined as 10°N to 10°S, which originated poleward of 20° latitude, both in total and individually from the northern and southern hemispheres. The results agree very well with the estimates from the tracers, and show that inner tropical air is not substantially diluted by isentropic import of mid-latitude air in only a shallow layer, from 580 to 680 K in March 1994 and from 530 to 590 K in October 1994. Outside this layer, the import of mid-latitude air is 40%-50% in the tropical upper troposphere and lower stratosphere with a tendency for the spring hemisphere to have been the major contributor. We may note that 1994 was a year with
a westerly phase of the quasi-biennial oscillation (QBO); the 1993 equivalent of Fig. 18, when the QBO was easterly in the lower atmosphere, showed a much weaker restriction near $\theta = 600$ K, with numbers of 10%-20% admixture of mid-latitude air rather than the $\sim 5\%$ in 1994. In the $300 < \theta < 550$ K range, the numbers were generally similar to those in Fig. 18, i.e. 25%-60%. With regard to the tropical trajectories, we remark in passing that the analysed (and forecast!) winds in the tropics were frequently in error, irrespective of whether ECMWF, UKMO or NMC information was used. An example is shown in Fig. 19(a) for the flight from Fiji to Hawaii on 941024, when the observed zonal components of the winds experienced by the ER2 were easterly at up to 35 m s$^{-1}$, as compared with analysed speeds of about half this; there were also very substantial errors in the meridional winds, see for example the traces for the Hawaii to Fiji flight of 940327 in Fig. 19(b). If the real atmosphere has larger meridional velocities than the analysed ones incorporated in the trajectories, then the percentages in Fig. 18 would be likely to increase. There are too few aircraft sampling days to make any statistically reliable statement.

We turn now to the examination of some individual profiles of water, NO$_x$ and N$_2$O, to make some points which are not evident from mean profiles, or which are clearer in individual observations. The first point is that the minimum in total water was co-located with the temperature minimum in both March and October over a wide range of locations, from Hawaii in March to Fiji in both March and October. This condition, coincidence of the tropopause and hygropause, was frequently observed near the equator and was also seen at 24$^\circ$S above typhoon 'Usha' on 940329, en route from Fiji to Christchurch. The hygropause was also at or below the tropopause on northbound flights from Christchurch on 940403, 940528 and 940801, at about 20$^\circ$S. Some of these vertical profiles of water, temperature, and N$_2$O are illustrated in Fig. 20. Thus drying at the tropopause is not restricted to the

![Figure 19.](image)

(a) Zonal wind components for the flight of 941024 from Fiji to Hawaii. The dashed line is measured velocity from the ER2, the solid line is analysed from the UKMO UARS analysis. Note the 35 m s$^{-1}$ easterly component near 7$^\circ$ N. (b) Comparison of meridional wind components for the flight of 940327 from Hawaii to Fiji. The dashed line is measured velocity from the ER2, the solid line is analysed from the UKMO UARS analysis. The aircraft was flying at $\sim 60$ hPa in both cases.
Figure 20. ASHOE profiles of water, temperature and nitrous oxide near the tropical tropopause as a function of potential temperature, illustrating the close proximity of the hygropause and the tropopause over a wide range of latitudes and times near the date-line. (a) Descent into Hawaii on 940320, (b) dip above typhoon 'Usha' at ~ 24°S on 940329, (c) dip at ~ 20°S, 173°E on 940801, and (d) descent into Fiji on 941022.

western tropical Pacific during northern winter; the effects were visible in the longitude sector from 155°W to 170°E whenever measurements were made between March and October from 10°N to 24°S, and over Hawaii during March. Examples for 940320 and 941022 are shown. Of course, the production of low water mixing ratios may still have been local and then spread more widely by advection.

A further interesting question in this regard is whether or not there is a thin layer of subvisible cirrus at or just below the tropical tropopause. There are no reliable data on particle size distribution in the tropics during March, because of instrumental difficulties.
Figure 21. Vertical profiles of particles and temperature, taken near the equator on 941026. For H$_2$O, NO$_x$, and N$_2$O, see Fig. 16. (a) Condensation nucleus counter (CNC) particle number density and temperature, (b) MASP particle number, surface and volume concentrations, and (c) mean CNC particle number density and total water relative to the tropopause. ASHOE phase 4 mean.

During October however, there are such measurements; there is one flight with clear evidence for a layer of very thin cirrus just beneath the tropopause, both near the equator and somewhat differently above Hawaii. The particle profiles from this flight, 941026, are shown in Fig. 21; the NO$_x$, H$_2$O and N$_2$O have been shown in Fig. 16. The enhanced layer of total water from 371 to 378 K correlates well with enhanced particle number, area and volume as measured by the Multi-Angle Aerosol Scattering Probe (MASP) which detects particles larger than about 0.02 µm. The condensation nucleus counter (CNC),
which detects all particles larger than 0.008 μm, shows a minimum where the MASP and the total water indicate a thin cirrus cloud. This does not accord with suggestions that such clouds produce aerosol particles; however, note that the minimum in total water at, and just above, the tropopause is correlated with a maximum in the CNC data but not in the MASP data. A possible inference is that the ice clouds which settled out to produce the dehydrated air, left behind new, small particles. Such a production mechanism for new particles is plausible, since in the saturated environment of a tropical thunderstorm top, any remaining condensable vapours (H₂O, H₂SO₄, HNO₃) would homogeneously nucleate once the ice crystals had sedimented out of the coldest air. Study of individual thunderstorm tops would be necessary to pursue such possibilities further. Figure 21(c) shows that the maximum number density in CNC aerosol particles coincides with the minimum in total water for the average of all phase 4 profiles in the inner tropics. Brock et al. (1995) have argued that binary homogeneous nucleation in the upper tropical troposphere is a source of condensation nuclei for the lower stratosphere.

The advection from mid latitudes is illustrated in Fig. 22. Not only is there laminar structure, with layers of older, N₂O-depleted air above the inner tropical tropopause near 395 K and 410 K, but also the relatively isothermal structure characteristic of the lower mid-latitude stratosphere is also present, rather than the sharply defined temperature minimum normally found. This isothermal structure cannot be the result of recent local deep convection because of the N₂O profile; once present, however, it could facilitate the penetration of cumulonimbus turrets to higher altitudes than would be the case with a more usual increase of temperature with height above the tropopause.

The issue of injection through the tropical tropopause of NOₓ produced by lightning may also be examined. Previous work (Murphy et al. 1993) has considered this thoroughly, using observations from the STEP*-'Tropical (Danielsen 1993) and AAOE (Tuck et al. 1989) missions. The work there will not be revisited, but we will consider scatterplots for tropical flights of NOₓ and N₂O such as those given in Fig. 2(c). Figure 23 shows composite scatterplots for the tropical flights in (a) March and (b) October. The feature we

* Stratosphere–Troposphere Exchange Project.
draw attention to here is the range of the NO$_x$ values at a given value of N$_2$O (Fig. 24). This range is larger for high N$_2$O values (recently injected air) than for low N$_2$O values (older air). The highest NO$_x$ values at a given N$_2$O mixing ratio are found in the northern tropics (7°N–20°N) while the lowest NO$_x$ values are found in the southern tropics (7°S–20°S). These wide distributions in high-N$_2$O air could arise either from the inherent variability likely to be associated with a lightning source, or from the variability of NO$_x$ in the troposphere as a result of HNO$_3$ being water soluble. We can examine this issue further by inspection of Table 6, which gives the value of NO$_x$ at the tropopause (note that we cannot use F(NO$_x$) (Tuck et al. 1994) because the function is ill-conditioned near the tropical tropopause, in part because of the recirculation of stratospheric air noted earlier). There is a systematic difference in the tropics during both March and October, with larger values at the tropical tropopause in the northern hemisphere than in the southern. Since there is no obvious reason why the ITCZ should produce more NO$_x$ in lightning in the northern tropics than in the southern tropics, particularly since it is largely south of the equator.


<table>
<thead>
<tr>
<th></th>
<th>37°N</th>
<th>21°N</th>
<th>14°N</th>
<th>0°</th>
<th>10°N</th>
<th>18°S</th>
<th>20°S</th>
<th>30°S</th>
<th>44°S</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Mean</strong></td>
<td>0.74</td>
<td>0.62</td>
<td>0.80</td>
<td>0.48</td>
<td>0.24</td>
<td>0.63</td>
<td>0.69</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Standard deviation</strong></td>
<td>—</td>
<td>0.31</td>
<td>0.25</td>
<td>0.29</td>
<td>—</td>
<td>0.09</td>
<td>0.52</td>
<td>—</td>
<td>0.23</td>
</tr>
<tr>
<td><strong>Mean</strong></td>
<td>0.34</td>
<td>0.74</td>
<td>—</td>
<td>0.42</td>
<td>0.38</td>
<td>0.44</td>
<td>—</td>
<td>—</td>
<td>0.90</td>
</tr>
<tr>
<td><strong>Standard deviation</strong></td>
<td>—</td>
<td>0.32</td>
<td>—</td>
<td>0.07</td>
<td>0.02</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>0.41</td>
</tr>
<tr>
<td><strong>Average over all phases</strong></td>
<td>0.54</td>
<td>0.67</td>
<td>0.80</td>
<td>0.44</td>
<td>0.38</td>
<td>0.31</td>
<td>0.53</td>
<td>0.64</td>
<td>0.82</td>
</tr>
<tr>
<td><strong>Standard deviation</strong></td>
<td>0.29</td>
<td>0.31</td>
<td>0.25</td>
<td>0.18</td>
<td>0.02</td>
<td>0.13</td>
<td>0.33</td>
<td>—</td>
<td>0.33</td>
</tr>
</tbody>
</table>

During northern winter, it is possible that some of the difference arises from the transport of higher NOx mixing ratios in the northern troposphere to the tropical tropopause. The difference is unlikely to arise from the horizontal advection from mid latitudes, since there were comparable and higher values of F(NOx) at 44°S in March and October, respectively, than at 37°N. This interpretation is consistent with the CO profiles in Fig. 17, where higher CO values are evident at the northern tropical tropopause, particularly during March. We may examine further the question of NOx injection through the tropical tropopause by means of scatterplots of it and of the total number density of condensation nuclei versus carbon monoxide. We have shown earlier that CO has an interhemispheric asymmetry not only in the troposphere, where there is more in the northern hemisphere because of extra industrial activity, but also in the stratosphere, presumably because the stratosphere of each hemisphere is fed predominantly by its own troposphere. Figure 25 shows scatterplots of CO versus particle number density for (a) March and (b) October. It is clear that during March the northern tropics are populated with high-CO, abundant-particle air, while the inner and southern tropics are populated by low-CO, scarce-particle air. In October the two populations join at low values of CO and particle number density; this occurs in regions of θ above about 450 K. However, while low-CO air is found throughout the tropics, there is a branch of high-CO air confined entirely to the northern tropics. Figure 25(c) shows that the condensation nucleus concentration has the same lower-stratospheric asymmetry, in a mean sense, between the hemispheres as CO (Fig. 17(c)), although it is less clear because of the peaked structure. The CN may reflect both the production at the tropical tropopause during dehydration (Fig. 21(c)) as well as a higher concentration in the northern hemisphere troposphere. The CO vs. NOx scatterplots in Fig. 26 show similar behaviour; for NOx values less than 2 p.p.b.v. in March, there is a separation between the hemispheres, a feature absent in October. This may have important consequences for the fractional abundance of NOy relative to N2O in the two hemispheres. Figure 27 shows the scatterplot of NOy vs. N2O for a flight in February north to the edge of the arctic vortex (60°N) over Canada with four flights in late March which cover the latitude range 21°N to 68°S. A flight on 941104 to 60°N over North America is also included for comparison of the autumn season between the hemispheres. The double structure at N2O values 250–300 p.p.b.v. has been discussed earlier in the context of Fig. 2(c); it arises during the flight from Hawaii to Fiji on 940327 with the upper, high-NOy branch occurring north of the equator. Note that the points on Fig. 27 south of 20°S form a population which curves toward high NOy values at low N2O (which are found at high latitudes near 68°S in the polar vortex). We may ask about the origin of the low-NOy air in the southern hemisphere relative to the northern hemisphere during late March in the tropics. Figure 25(a) shows that the asymmetry in CO between the hemispheres extends over the entire latitude range, 60°N to 70°S, in March. The lower
Figure 25. Scatterplots of CO vs. total particle number density from the condensation nucleus counter (CNC). Note the hemispheric asymmetries. (a) Phase 1, (b) phase 4, and (c) latitudinal profiles of CNC, 60°N to 70°S during ASHOE phases 1 and 4; cf. Fig. 17(c).

Figure 26. Scatterplots of CO vs. NO_x, flights as indicated, (a) March and (b) October.
Figure 27. NO$_y$ vs. N$_2$O scatterplot for five flights in 1994, covering the latitude range 21°N–68°S. One point in 20 is shown in (a). Note the relatively low NO$_y$ in the tropics, and the curvature of the high southern latitude points (generally low N$_2$O) towards higher NO$_y$. (b) The points near the equator in greater detail. The southern hemisphere (SH) March data in (a) are different from the northern hemisphere (NH) whether the latter were taken in February, March or November; the SH October data (not shown) are like the NH data. It is suggested that the interhemispheric difference in March originates mainly in the tropics during northern winter.

stratospheric air in mid-latitude air therefore retains a signature of tropical air, but it is mixed with air which has descended at higher latitudes, as is clear from the N$_2$O traces in Fig. 6. The separation between the northern and southern branches in Fig. 27 stabilizes at ~1 p.p.b.v. of NO$_y$ at 180 < N$_2$O < 285 p.p.b.v., decreasing at lower, higher latitude values of N$_2$O. Approximately half of this can be accounted for by the difference in NO$_y$ at the tropical tropopause either side of the equator (Table 6, Fig. 24). The remaining half a p.p.b.v. could be accounted for in two ways: (i) the transport of air from the outer arctic vortex enriched in NO$_y$ by PSC particles falling from aloft (Hübler et al. 1990; Kawa et al. 1992a) or (ii) it could be a memory from the denitrification of the arctic vortex which broke up in November 1993 during the 1993/94 boreal winter (Keim et al. 1996, personal communication). It seems unlikely that a signature of low NO$_y$ from the break-up of the arctic vortex would persist over the entire southern hemisphere for 4–5 months; a signature of relatively high NO$_y$ in the northern hemisphere from sedimentation in PSCs followed by evaporation in the outer arctic vortex in the first two months of 1994 seems more probable, possibly accompanied by simultaneous injection of relatively high NO$_y$
air in tropical thunderstorms, a process probably at its peak in northern winter. Note that the difference between the hemispheres is present in Fig. 27 whether the comparison is made with the March or November values in the northern hemisphere. On the flight north to the edge of the arctic vortex on 940218, there is evidence of both denitrification and enhancement by evaporation of particles falling from aloft in the vertical profile there (not shown), and evidence for relatively high NOx in the ‘young’, high-N2O air near 37°N. We shall return to this subject in section 5, on mid latitudes.

4. THE DOWNWARD BRANCH: VORTEX

(a) Horizontal structure

We will begin by showing the evolution of the mean horizontal wind profile observed from the ER2 at high southern latitudes from late March to October near the date-line, during the four phases of the ASHOE/MAESA. In each phase the mean latitude of the wind speed maximum was at ~ 66°S, with variations caused by synoptic variability of up to 15° latitude. Figure 28 shows that the maximum wind speed at the jet core increased from a mean of 25 m s⁻¹ in late March/early April to 35 m s⁻¹ in late May and early June. In late July/early August and in October, the mean jet maxima were, respectively, 52 and 53 m s⁻¹, with the latter having steeper wind shear, particularly on the anticyclonic side. The fact that the jet core was at the same average latitude during all four phases makes it a natural definition of the vortex boundary; areas bounded by any selected PV contour grew until mid winter, remained constant for several weeks and then decreased, just as they did in 1987 (Tao and Tuck 1994). We further note that scatterplots of operationally analysed PV versus observed tracers were in general far from compact, as found by Tao and Tuck for 1987. An example is shown in Fig. 29, and it is clear from it that techniques which use analysed PV to calculate tracer distributions will have limited accuracy: during this flight, the PV could explain less than half of the variance in the N₂O, and this approach also loses the phase information. Note that the filaments of ex-vortex air (characterized by low values of N₂O) from 63°–65.5°S, 56°S–62°S and in diluted form at 51°S and equatorward of 46°S are not well matched by PV. The reasons for this may be seen in Fig. 30, where the observed and analysed wind speeds and directions are compared. There is much mesoscale structure in the observations which is not in the analysed wind profile. The fractional N₂O

![Figure 28. Mean horizontal wind speed, centred on an origin at the jet core, for phases 1–4 of the ASHOE. The mean position of the jet core was at about 66°S in each case.](image-url)
Figure 29. Comparison of N$_2$O and potential vorticity, PV, (UKMO UARS analysis) for the flight of 940603, which was due south from Christchurch to 67ºS and back. Note in (a) that the N$_2$O vs. PV scatterplot is not compact, and in (b) that the PV vs. N$_2$O relationship reproduces the observed N$_2$O along the flight track poorly, even with respect to the larger-scale alternations of low and high mixing ratio.

Figure 30. Comparison of observed and analysed (UKMO UARS) winds for the flight of 940603: (a) wind speed and (b) wind direction. Note the structure on all scales in the observations, and the velocity discrepancies at the southern end of the flight track. Dots, ER2 observations; solid, UKMO UARS analysis.

variance explicable by the analysed PV varied from flight to flight, sometimes being less than 20% and sometimes more than 80%. An average was in the 40%–60% range, with better performance during phases 3 and 4 than in phase 2. The analysed PV had difficulty in positioning filaments accurately, with errors of several degrees of latitude being common even with large, single structures. For these reasons, where some measure of the location of the vortex edge is required, we will use the observed wind maximum. The vortex edge is not a concept which will bear too much formal definition; there are frequently multiple ‘edges’—sharp horizontal gradients in tracers—as may be seen in Fig. 31 for flights in early August and early October, so the use of a single edge in horizontal tracer gradients to define the vortex is ambiguous. Such multiple edge structures show the broad polar-night jet to be a region of horizontal exchange. We may also note that on one flight, 940605, the 4-hour southbound leg and the 4-hour northbound leg were, other than in the dip, separated by an average ≲ 5 K in potential temperature, and yet there is poor correlation of the tracer.
structures on the two legs, implying that at least on this day they were shallow, short-lived or both (Fig. 32). On other flights, however, there were on occasion similar structures on both legs in regions where the vertical separation of the flight tracks approached $\Delta \theta \approx 40$ K, particularly during October.

It was suggested from the AAOE mission (Murphy et al. 1989) that there was an exchange zone $\pm 4^\circ$ latitude either side of the edge of the region of high ClO and low ozone, based on the behaviour of the correlation coefficient between these two species. Pierce et al. (1994) concluded that there was a zone $10^\circ$ latitude wide centred on the jet maximum which was a mixing zone, modulated by the effects of diabatic descent, based upon studies of 'chaotic advection' (exponential lengthening of material filaments over time) and satellite observations. Here we look at the seasonal behaviour of this zone, based upon measurements of the fractional abundance of HCl, NO$_x$ and water between late May and late October. All three of these species are ones which decrease in abundance as a result of processes occurring in the winter antarctic vortex. HCl was lost by heterogeneous chemical processing when temperatures dropped below 195 K (Tuck et al. 1995a), whereas loss of nitric acid, HNO$_3$, and water occurs by gravitational sedimentation of particles large enough (diameter $d \geq 4 \mu$m) to fall appreciable distances (km) on the time-scale of a day. Such sedimentation appears to demand temperatures at frost-point or colder, with evidence from the arctic that denitrification can occur in the absence of appreciable dehydration. Thus, as the antarctic vortex cooled in 1994, temperatures at $\sim 50$ hPa first reached 195 K in May, and frost-point ($\sim 188$ K) in June. There is also a tendency for
Figure 32. Northbound and southbound legs for the ASHOE flight on 940605. Both legs were flown nominally at potential temperature $\theta = 445$ K, and the mean separation in $\theta$ was less than 5 K. In the course of the 8-hour flight, the small-scale features changed, and the sharp $N_2O$ gradients near 62°S and 48°S on the southbound leg had moved equatorward by the time they were encountered on the northbound leg. Note that the $\approx 20$ p.p.b.v. difference in $N_2O$ between 54°S and 61°S greatly exceeds what would be obtained from multiplying $\Delta \theta$ over that stretch by the mean $N_2O$ lapse rate: it is a temporal change induced by horizontal advection. Note also the low–high–low–high pattern of $N_2O$ between 45°S and 70°S.

Figure 33. Fractional abundance of HCl, calculated relative to Cl$\nu$ from Sala\'witch's formula, along the ASHOE flight tracks of 940524, 940601, 940603 and 940605. The fractional abundance of HCl dropped rapidly both inside and outside the vortex after 940601. The highest values occur in air of recent low-latitude origin (see Figs. 29 and 32 for 940603 and 940605 $N_2O$ traces).

the coldest temperatures to be found well inside the vortex, with a bias towards West Antarctica. Figure 33 shows the fractional abundance of HCl along ER2 flight tracks, relative to the wind speed maximum, for a sequence of 4 days: 940524, 940601, 940603 and 940605. Poleward of the wind maximum (i.e. inside the vortex) loss of HCl occurred during the 8-day period between 940524 and 940601 (up to 35%). This loss reached 60% by 940603, and during the period 1–5 June there was substantial HCl loss, ranging up to 30%, outside the vortex. Since the requisite temperatures occurred largely poleward of the wind maximum (Tuck et al. 1995a, b), most of the effect arose from peel-off of low-HCl air from the vortex, as seen in the $N_2O$ traces in Figs. 29 and 32. There was, however, no concomitant loss of NO$_x$ or H$_2$O observed from the ER2 in June, and nor was such loss seen in late July or early August other than on an intermittent basis inside the vortex. It was
Figure 34. Medians of $R(H_2O)$ from (a) the AAOE, August–September 1987 and (b) phases 1–4 of the ASHOE/MAESA. In (a) the curves are for potential temperature $\theta = 425 \pm 10$ K and $\theta = 450 \pm 10$ K. In (b) all curves are for $\theta = 430 \pm 10$ K.

clearly seen in early October however, but not to such an extent as was seen in the South American sector in late August and September 1987, either inside or outside the vortex. This may be seen by comparing the residual water, $R(H_2O)$, and the fractional NO$_x$ abundance relative to N$_2$O lost, F(NO$_x$), for the AAOE and ASHOE/MAESA (for definitions see Tuck et al. (1994)). Residual water is a measure of the average water vapour content of air entering the stratosphere; it is derived from simultaneous stratospheric observations of water vapour and methane by subtracting the water produced as a result of methane oxidation in the stratosphere. Its value has been observed to be in the range 3–4 p.p.m.v. on previous missions, except where polar dehydration had lowered the value. Figure 34 shows the medians of $R(H_2O)$ from the AAOE, relative to the jet-stream maximum at potential temperatures of 425 $\pm$ 10 K and 450 $\pm$ 10 K. Figure 34 also shows equivalent observations for the ASHOE/MAESA, at potential temperatures 450 $\pm$ 10 K, for all four phases. The equivalent data for F(NO$_x$) are shown in Fig. 35. It is apparent that the abundance of water in the South American sector during August–September 1987 was considerably less than it was at any time between March and October 1994 in the New Zealand sector, whether the comparison is made inside or outside the vortex. It is moreover apparent that there is a pronounced increase in F(NO$_x$) from early April to early June during 1994, with a similar but much lesser tendency in $R(H_2O)$. The behaviour for both F(NO$_x$) and $R(H_2O)$ from mid winter to October is more variable. We will examine the question as to whether the year or the longitudinal sector is responsible for the AAOE–ASHOE/MAESA difference in $R(H_2O)$ later. The question as to why F(NO$_x$) shows a seasonal increase whereas $R(H_2O)$ does not is somewhat more easily addressed. The vertical profiles measured near 67°S (i.e. near the polar-night jet core) for phase 1 during late March/early April are shown in Fig. 36.
Figure 35. Medians of F(NO$_2$) from (a) the AAOE and (b) phases 1–4 of the ASHOE/MAESA. Potential temperature, θ, intervals are as in Fig. 34; note that all flights where PSC-enhancement of the NO$_2$ measurements could have occurred have been excluded.

R(H$_2$O) is vertical, having no lapse rate, whereas F(NO$_2$) displays a positive lapse rate, increasing from 0.052 at θ = 415 K to 0.061 at θ = 485 K, an 18% increase in about 3.5 km. It follows, therefore, that the diabatic descent which occurs throughout autumn, winter, and spring will increase F(NO$_2$) at ER2 altitudes but leave R(H$_2$O) unaffected, in polar regions. We have summarized the seasonal variation in 1994 for 5° latitude belts relative to the jet-stream core for N$_2$O, NO$_2$, F(NO$_2$) and R(H$_2$O) in Fig. 37. The observations are all those taken in horizontal flight in the range θ = 450 ± 10 K. From the N$_2$O curves, several points are apparent. For the latitude belt 5° to 10° poleward of the jet core, the mean mixing ratio declined from 230 p.p.b.v. in late March/early April to 115 p.p.b.v. in October. The belt 0° to 5° poleward of the jet core declined from 220 p.p.b.v. in phase 1 to 150 p.p.b.v. in phase 4. This indicates two things: that there is a mean radial gradient in the outer vortex, and that the evolution over the 7-month winter has been such that the balance between the meridional mixing (which maintains the high end of the radial gradients) and the continuing diabatic descent (which maintains the supply of low-N$_2$O air from aloft) is such that there has been a net lowering of the N$_2$O isopleths. The four belts within 10° of the jet core all showed a similar pattern; the decrease was not monotonic—the N$_2$O showed an increase during phase 3. This increase indicates that the tracer content of the outer vortex is a dynamic balance between horizontal mixing and diabatic descent; an increase in phase 3 cannot otherwise be accounted for, even if dθ/dt had become zero—which was not the case, as we shall see later. Note that it is possible only to get a lower limit for the diabatic descent rates by considering this lowering of the N$_2$O isopleths, a fact which limits the value of time series of profile observations from a single antarctic station, such as those for N$_2$O and H$_2$O respectively analysed by Crewell et al. (1995) and
Figure 36. Mean vertical profiles of potential temperature vs. R(H₂O) and F(NOₓ) near 67°S for phase 1 of the ASHOE/MAESA.

Vömel et al. (1995); we will discuss values of d½dt estimated directly from high-resolution interferometric radiometer measurements later. The behaviour of the N₂O curves further equatorward of the jet core is interesting. The belt centred at 12.5° equatorward of the jet core (i.e. at about 55°S) showed N₂O decreases from phases 1 through 3, with an increase in phase 4 (October). The belt centred 17.5°S equatorward of the jet core had minimum values of N₂O during phase 2 (late May/early June). This suggests that during phase 2, when the jet was only about 2/3 of its eventual strength (Fig. 28) but when nevertheless there was appreciable HCl conversion to reactive chlorine by PSC processing, there is significant transport of processed vortex air to mid latitudes, as may be seen in Fig. 33 for specific examples. One feature of Fig. 37 is that the curves for N₂O sometimes cross, as do those for NOₓ, F(NOₓ), H₂O and Hₓ (the ratio of HCl to total inorganic chlorine). This is likely to be a reflection of the high-low-high structure seen in tracers along flight tracks (Fig. 32) and on PV maps. There was no penetration of the full payload into the 5°–10° belt poleward of the jet core during phase 4, and this accounts for the absence of a denitrification signal equivalent to the dehydration then. The most equatorward belt in F(NOₓ) behaves differently from the others, and probably marks the changeover between belts more influenced by the vortex and those more influenced by the tropics; it is centred 17.5° of latitude outside the jet core. It seems that there is a smaller increase in F(NOₓ) outside the vortex between phases 3 and 4 than might have been expected; a possible explanation is some communication of denitrification across the jet to mid latitudes—the denitrification is evident in the 5° belt immediately poleward of the core. We will return to the behaviour of F(NOₓ) later, and in the following section on vertical structure. Residual water, with its absence of a vertical gradient, shows the effects of horizontal motion more visibly; there are increases between phases 2 and 3 followed by decreases between phases 3 and 4, particularly in the five most poleward belts. This is most likely due to enhancement of water by evaporation of ice crystals falling from above in mid winter followed by the spread of vortex dehydration in late winter and early spring. The abundance of Hₓ drops in
all belts excepting the most equatorward up to and including phase 3, when if conserved it would have increased, since the CFCs are highly correlated with N$_2$O. The behaviour of HCl between phases 3 and 4 inside the vortex is consistent with the recovery under the uniquely antarctic vernal conditions of 90% ozone loss (Austin et al. 1989; Prather and Jaffe 1990).

Most analyses of the containment or spread of antarctic denitrification and dehydration have started with the assumption that once the condensable vapours have been lost, there is no resupply, and that the sedimentary particles fall out of the stratosphere. Tuck et al. (1994) discussed the possibility that in the outer antarctic vortex the falling particles could evaporate in the stratosphere, because the temperature profiles there frequently show a layer of warm air in the stratosphere beneath the regions saturated with respect to ice. Evidence of such behaviour was seen in the arctic (Hübner et al. 1990; Kawa et al. 1992a) but has not been reported in the South American sector of the antarctic in 1987, although there is evidence that denitrification and dehydration were separate processes there too (Tuck et al. 1994). We will now examine observations from flights in early June, early August and October which show that in the New Zealand sector in 1994 both NO$_x$ and water displayed evidence of enhancement in the outer vortex, often at levels well above
Figure 38. NO\textsubscript{y} vs. N\textsubscript{2}O scatterplots for four southbound flights from Christchurch. On none of these flights was there any evidence for a PSC encounter. (a) 940601, (b) 940806, (c) 941013, and (d) 941016. Low-NO\textsubscript{y} deviations from the compact, near-linear curve indicate denitrification, high ones indicate probable enhancement by evaporation of HNO\textsubscript{3}-containing particles falling from aloft. The high-NO\textsubscript{y} deviations on 940806 are inside the vortex and above potential temperature $\theta = 420$ K. The high-NO\textsubscript{y} deviations on 941013 and 941016 are outside the vortex.

$\theta \approx 400$ K. Furthermore, in October this signature was found outside the vortex. We also re-examine the AAOE observations from 1987.

Figure 38 shows the scatterplots of NO\textsubscript{y} vs. N\textsubscript{2}O for four flights: 940601, 940806, 941013 and 941016. The deviations from the near linear, compact plot on the high NO\textsubscript{y} side signify the enhancement of NO\textsubscript{y} by the evaporation of particles falling from above. Note that the effect was seen as early as 940601, was evident above $\theta \approx 400$ K on 940806, and at $500 < \theta < 520$ K outside the vortex on 941013 and 941016. There was an absence of PSCs anywhere on these flights, precluding the possibility of artefactual NO\textsubscript{y} enhancement by the anisokinetic sampling of type I PSC particles (Fahey et al. 1989). The flights of 940806 and 941013 also show denitrification—in the outer vortex—via the populations on the low NO\textsubscript{y} side of the main linear anticorrelation.

Armed with knowledge that vertical redistribution of a condensable vapour—HNO\textsubscript{3}—occurred, we will search for evidence of similar behaviour by water vapour, recalling that dehydration may be a separate process from denitrification even over Antarctica (Tuck et al. 1994). The fraction of water vapour in the lower stratosphere which arises from methane oxidation is rather low (less than $\approx 30\%$ on average), unlike the fraction of NO\textsubscript{y} arising from N\textsubscript{2}O photolysis, which is typically more than $\approx 85\%$. It follows that CH\textsubscript{4} vs. H\textsubscript{2}O scatterplots will be less compact than those for NO\textsubscript{y} vs. N\textsubscript{2}O, and will bear a larger signature of variability arising from the injection process at the tropical tropopause. We therefore categorize the CH\textsubscript{4}–H\textsubscript{2}O relationship by dividing each flight into segments, each of which represents a distinguishable air mass via its CH\textsubscript{4} content. We will consider four
Figure 39. \( \text{H}_2\text{O} \) vs. \( \text{CH}_4 \) correlations for 940605. The ASHOE flight is divided into 19 separate air masses, based upon the \( \text{CH}_4 \) trace, and a correlation is worked out for each. Note the generally consistent negative correlation for each air mass and overall.

Figure 40. \( \text{H}_2\text{O} \) vs. \( \text{CH}_4 \) correlations for 940808. Segments 17 and 18, which are outside the vortex, are deficient in water vapour, while segments 10–12 inside the vortex contain moister air than would be expected from their methane content. Both features probably arise from the sedimentation of ice crystals, with evaporation at flight level before falling from the stratosphere in the latter case.
Figure 41. \( \text{H}_2\text{O} \) vs. \( \text{CH}_4 \) correlations for 941003. Segments 9–11 show the vortex dehydration. There is enhancement in segments 12–14.

Figure 42. \( \text{H}_2\text{O} \) vs. \( \text{CH}_4 \) correlations for 941016. Segments 6–9, 12 and 18–19 show dehydration well outside the vortex, and possible enhancement in segments 1–5.
flights, 940605, 940808, 941003 and 941016. The first of these shows a CH$_4$–H$_2$O relation (Fig. 39) which is similar for all 19 air masses into which the flight has been divided. The slopes are generally between 0 and $-2$, with a mean value close to $-1$. This does not mean that one water molecule is produced per methane molecule oxidized; rather it reflects the averaged effect of temporal and spatial variations imposed on water vapour at the tropical tropopause upon entry to the stratosphere, combined with the effects of methane oxidation. The 940808 flight is shown in Fig. 40. It is apparent that the 18 different air masses exhibit a different picture than their counterparts did 2 months previously; the air in segments 17 and 18, which are at high potential temperatures ($\theta > 500$ K) outside the vortex, is deficient in water vapour content, while that in segments 8–12 contains some air which is richer in water vapour than would be expected from its methane content. The obvious interpretation is that the flight encountered both dehydrated air and air enhanced in H$_2$O by evaporation of ice crystals falling from above. Note that such processes have the potential to obliterate any simple correlation between H$_2$O and CH$_4$; they will also limit attempts to compute time constraints for the flow through the vortex using the polar dehydration and denitrification. We also consider these signatures in the 941003 and 941016 flights. Figure 41 shows the dehydration inside and at the edge of the vortex on 941003 (segments 9–11), and also some enhancement (segments 10–14). Note that the minimum 50 hPa temperatures over Antarctica in 1994 did not exceed 188 K, the likely frost-point, until 941019 (Tuck et al. 1995a), much later than the mid-September date suggested by Vömel et al. (1995). The H$_2$O vs. CH$_4$ scatterplot for 941016 shows dehydration well outside the vortex at $\theta > 480$ K (segments 6–9, 12 and 18–19) and perhaps some enhancement in segments 1–5 (Fig. 42). The dehydration exterior to the vortex above 480 K also shows depleted ozone, as seen in the O$_3$ vs. N$_2$O scatterplot in Fig. 43, providing evidence of transport of polar ozone loss to mid latitudes.

The 1994 results just described suggest that it would be worth re-examining the 1987 (AAOE) observations for signs of NO$_y$ and water enhancement via evaporation of particles falling from above. The data are presented in the form of medians from all qualifying flights. Because the NO$_y$ and water instruments experience increased signals when particles from respectively type I and type II PSCs enter their inlets (Fahey et al. 1989; Kelly et al. 1989), we exclude any AAOE flights on which the relevant PSCs were present. This led to the exclusion of half the flights for F(NO$_y$), (870817, 870818, 870828, 870830, 870902, 870904) when type I PSC activity was seen in horizontal flight (Pueschel et al. 1989; Ferry et al. 1989) and of 870817 and 870818 for R(H$_2$O) when type II particles were seen. The variability of F(NO$_y$) and R(H$_2$O) along the flight tracks was such that medians were more useful than means, although essentially the same results were obtained in each case. Before displaying the medians, however, we show an individual flight which illustrates some important features. The trace of F(NO$_y$) at $\theta = 425 \pm 10$ K is shown for 870909 relative to the jet-stream core in Fig. 44. At a position of $2^\circ$–$5^\circ$ inside the core there is a peak in F(NO$_y$), in particle-free, air, reaching a maximum of 0.075. There is no corresponding dip in N$_2$O, so the F(NO$_y$) peak cannot be the result of simple gaseous descent; particle evaporation is the likely explanation. Excursions to high values of F(NO$_y$) at or outside the jet core can be seen on several flights, as can low F(NO$_y$) filaments presumably peeled off from the denitrified part of the vortex. The medians show, at both $\theta = 425 \pm 10$ K and $\theta > 440$ K, a region of enhanced F(NO$_y$) stretching across the 10° latitude wide belt centred on the jet core (Fig. 45). On the same diagram the medians for N$_2$O are shown; a ‘mixing slope’ across the core is evident, plus a maximum situated 5° to 10° of latitude equatorward of the jet core. This maximum may be a manifestation of the pattern seen on PV maps, with low absolute PV (high N$_2$O) air close to the vortex edge, and peeled-off vortex-edge air equatorward of it. There is no sign of a depression.
in N$_2$O across the jet core, which would be necessary to explain the high F(NO$_y$) belt by gaseous diabatic descent. There is a similar pattern for R(H$_2$O) at $\theta \geq 440$ K (Fig. 46), but not at $\theta = 425 \pm 10$ K, where there is a mixing slope similar to that of N$_2$O. A consistent interpretation is that while there is NO$_y$ enhancement in both $\theta$ layers, it occurs only at the higher level for water. Note that the absolute values of R(H$_2$O) are lower by 1 to 1.5 p.p.m.v. during the AAOE than the ASHOE/MAESA, while those for F(NO$_y$) are similar. Denitrification and dehydration are separate events, with the latter occurring first as temperatures approach frost-point; the observations suggest that water in the outer vortex may be enhanced at $\theta \geq 440$ K by evaporation of ice crystals falling from above, and NO$_y$ at all levels accessible to the aircraft. This interpretation, combined with the observation that PSCs of all types have their maximum frequency over West Antarctica, i.e. in the South American sector (Watterson and Tuck 1989), raises the possibility that the differences seen between the AAOE (1987, South American sector) and the ASHOE/MAESA (1994, New Zealand sector) in water vapour and residual water arise at least in part not from a temporal difference but a longitudinal one. Water vapour is lost over West Antarctica, and the dehydrated air peeled off there at $\theta = 425 \pm 10$ K, to mid latitudes, while higher up the crystals evaporate by falling into warmer air en route to East Antarctica. In this manner a
Figure 45. Medians of $F(\text{NO}_2)$ and $N_2O$ for the AAOE, relative to the wind speed maximum, for non-PSC I flights. (a) Potential temperature $\theta = 425 \pm 10$ K, and (b) $\theta \geq 440$ K. Note the enhanced $F(\text{NO}_2)$ across the $10^\circ$ latitude-wide belt centred on the jet core, in contrast to the 'mixing slope' shown by the $N_2O$.

Figure 46. Medians of $R(\text{H}_2\text{O})$ for the AAOE, relative to the wind speed maximum. (a) Potential temperature $\theta = 425 \pm 10$ K, and (b) $\theta \geq 440$ K. Note the enhanced $R(\text{H}_2\text{O})$ across the core of the jet in (b).
Figure 47. $\text{N}_2\text{O}$ and $\text{O}_3$ traces relative to the wind speed maximum for ASHAE flights on (a) 940603, and (b) 940806. Note the ozone loss which developed poleward of the jet core between the two flights.

A longitudinal gradient in $\text{H}_2\text{O}$ and $\text{R(H}_2\text{O})$ could be maintained, with low mixing ratios in the western hemisphere inside and outside the vortex. Such a distribution cannot be proven by a single aircraft, and will need to be demonstrated by satellite observations (Rosenlof et al., personal communication).

The peel-off of low-ozone air from the vortex seen on 941016 (Fig. 43) prompts the question when this might have first occurred. Since we have seen that there was evidence of the peel-off of PSC-processed air in early June, there could be mid-latitude ozone loss induced that early. The peeled-off filaments on 940603 were, however, high in ozone (Figs. 29 and 47); the ozone increased as the aircraft flew southward across the jet, because the high ozone mixing ratios had not then been attenuated by the elevated level of $\text{CIO}$ induced by the PSCs. Figure 47 shows traces of $\text{N}_2\text{O}$ and $\text{O}_3$ for 940603 and 940806, relative to the wind speed maximum. It is seen that on the earlier flight, $\text{N}_2\text{O}$ and $\text{O}_3$ are negatively correlated throughout, including the region poleward of the jet core. The $\text{CIO}$ there was $\approx 600\ p.p.t.v.$ (parts per trillion $10^{12}$ by volume) (Tuck et al. 1995a, b), and had reached $\approx 800\ p.p.t.v.$ by 940806. By the time of this latter flight, 2 months had elapsed, and the ozone poleward of the jet core was now positively correlated with $\text{N}_2\text{O}$, equivalent to $\approx 25\%$ loss at the vortex edge, where sunlight was available to drive the photochemistry even in midwinter, since the jet core encompasses a larger area than the terminator of the polar night, and since synoptic-scale distortions continually take the outer vortex air to latitudes as far equatorward as $50^\circ\text{S}$. Such distortions also induce peel-off (Tuck 1989; Tuck et al. 1992, 1994; Waugh et al. 1994).

Further evidence for mixing at the vortex edge in midwinter may be deduced from the behaviour of 10-day back trajectories from flights in early August. Tuck et al. (1995a,
b) showed that the recovery time of the fractional HCl abundance in the outer vortex from PSC processing could not be explained photochemically, but was the result of resupply of unprocessed air by mixing at the vortex edge. Such mixing may be seen in Fig. 48; trajectories there show all possible behaviours: processed inside and staying inside, processed inside and moving outside, processed outside and staying outside and even processed outside and moving inside. The first of these was the predominant behaviour, with the second and third being significant.

The degree of mixing implied by the above processes is considerable, since the 10° latitude belt centred at 67° latitude contains more mass than the entire cap poleward of it. This degree of transport 'out of the vortex' has been objected to on the basis of model calculations. Such models use radiative-transfer calculations which give diabatic cooling rates in the vortex $d\theta/dr$ smaller than 0.5 K day$^{-1}$. Here we examine the cooling rates deduced from the upwelling irradiances measured by the HIS instrument (Smith 1991), spectrally resolved with 0.5 cm$^{-1}$ resolution from 3.4 to 16.7 µm. This instrument made five successful flights during the ASHOE/MAESA; we will examine results from one of them, 940808, which achieved a deep penetration of the vortex. Figure 49 shows the temperature structure and cooling rates for 940808; two important features are apparent. The first is that there is a tongue of relatively warm air in the range 350 < $\theta$ < 500 K (200 to 50 hPa) which is apparent over the entire latitude range 45°S to 68°S; the jet core at $\theta \approx 450$ K was at 57°S. This warm tongue is not reproduced well either by the TOVS* observational system (Smith et al., personal communication) or meteorological analyses, see Fig. 50. Figure 50 shows UKMO analyses, the operational version of which tended to be marginally better than the others in this respect on average; none of the available analyses performed well, however. The warm tongue was seen by the HIS but poorly represented in analyses on all five flights (940413, 940808, 940810, 941003 and 941005).

* TIROS (Television Infra-Red Observation Satellite) Operational Vertical Sounder.
Figure 49. (a) Temperatures (K) and (b) cooling rates, $d\theta/dt$, (K day$^{-1}$), deduced from the HIS observations on 940808.

The cooling rates calculated from the HIS are the second important feature in Fig. 49. At $\theta \approx 450$ K, the value of $d\theta/dt$ is close to $-1.8$ K day$^{-1}$ along the whole latitude range, the most poleward 11° of latitude of which are inside the vortex. The cooling rates range from $d\theta/dt \approx -1.0$ K day$^{-1}$ well inside the vortex near $\theta \approx 400$ K (the base of the ‘restrained’ region of the vortex) to $d\theta/dt \approx -2.0$ K day$^{-1}$ near $\theta \approx 450$ K and just inside the jet-stream core. A conservative extrapolation to $\theta \approx 500$ K gives cooling rates there of 3 K day$^{-1}$ in $\theta$. The values between the tropopause at $\theta \approx 300$ K and the base of the restrained vortex at $\theta \approx 400$ K range from $d\theta/dt \approx -0.2$ K day$^{-1}$ at $\theta \approx 320$ K to $d\theta/dt \approx -0.85$ K day$^{-1}$ in the range $350 < \theta < 400$ K. These values are typical in general for the five flights, but
much research remains to be done on these observations, particularly as regards the relative dependence of the cooling rates upon the vertical temperature structure and the amounts of O$_3$ and H$_2$O. We note that calculated solar-heating rates along the flight track in Fig. 49 are considerably smaller, ranging from less than 0.1 K day$^{-1}$ at low $\theta$ and high latitudes to 0.25 K day$^{-1}$ at high $\theta$ and low latitudes. The resulting net cooling rates $d\theta/dt$ are still in the range 1–3 K day$^{-1}$ over 400 < $\theta$ < 500 K. Figure 51 shows the relative contributions to the cooling rates of the water vapour rotational band, CO$_2$ and ozone. As pointed out by Clough et al. (1992) and Sinha and Harries (1995), for low-temperature atmospheres, the water vapour rotational band needs careful representation. At the temperatures seen on 940808 in the antarctic vortex, the Planck black-body function is shifted so that much of the energy being radiated upward is being emitted by the water vapour rotational band on the long-wavelength side of the 15 $\mu$m CO$_2$ band. The calculation used to derive Figs. 50 and 51 was performed with MODTRAN3, validated against the line-by-line calculation of Clough et al. (1992), and using ER2 water vapour and ozone measurements in the stratosphere, and those retrieved by the HIS in the troposphere. The magnitudes of $d\theta/dt$ are considerably larger than those published from radiative-transfer calculations in freerunning numerical models, and are therefore capable of sustaining a larger flow (by about a factor of two to three) through the vortex than such models. A further interesting point in
regard to the warm layer observed by the HIS is that it will permit more ready evaporation of particles containing condensed HNO₃ and/or ice which fall into it from above, something which supports the interpretation of the NO₃ and H₂O observations offered previously.

(b) Vertical structure

The occurrence of laminae in the vertical profiles of chemical species, often correlated with similar structures in wind speed and direction, have been shown previously for both the antarctic in the South American sector and for the arctic in the Scandinavian sector (Tuck 1989, 1994). Vaughan and collaborators have demonstrated that the maximum frequency of occurrence of such laminae in the world-wide ozonesonde observations is at the vortex edge in winter and spring, between the tropopause and θ ≈ 500 K (Reid and Vaughan 1991; Reid et al. 1993; Reid et al. 1994). These may arise from inertia–gravity waves of short vertical and large horizontal wavelengths (Danielsen et al. 1991) or they may be the result of differential advection (Reid and Vaughan 1991; Orsolini 1995). It was possible during the AAOE and the AASE that there was a role of topography in the genesis of such laminae, although some were upwind of the antarctic and Scandinavian peninsulas; from New Zealand during the ASHOE/MAESA, however, the flights were entirely over the Southern Ocean from 47°S to 70°S. One or more laminae of varying amplitudes were
Figure 52. Vertical profiles of wind speed, wind direction and ALIAS CH₄ for 940808. The ASHOE profile was taken about 10° latitude poleward of the wind maximum; similar laminar structures were seen on the other profile taken well inside the vortex on 941003.

Figure 53. Vertical profiles of wind speed, wind direction and ALIAS CH₄ for 941010. The ASHOE profile was taken near the jet core; the laminar structures are a common occurrence in such a location, and are very variable, sometimes being limited to a single event in a profile.
observed on every southbound flight from Christchurch, in dips performed at 67 ± 3°S. The laminae were present both near the vortex edge, when the jet core was near 67°S, and well inside the vortex, when the jet core was in the 55°–60° latitude range. As on previous missions, the laminae were more frequent at θ ≤ 400 K. These features are illustrated in Figs. 52 and 53 for profiles, respectively, near the edge of, and well into, the vortex. Such structures are unlikely to be reversible and almost certainly involve exchange between the vortex and its environs. It is important to recall that ozone loss in the vortex is at a maximum below θ ≈ 400 K (Proffitt et al. 1989b; Murphy 1991), the region where the laminae are most frequent and where trajectory analyses and PV maps show the exchange with mid latitudes to be the least restrained.

The vertical structure of the expressions F(NO₂) and R(H₂O), which represent the behaviour of the species which can condense within the vortex, changed over the course of the 1994 austral winter. Figure 54 shows that above θ ≈ 410 K, F(NO₂) increased throughout the March–October period at 67°S. Note that although the slope of the vertical profile was positive during phase 1, it was almost zero during phase 2 before coming positive again during phases 3 and 4. Gaseous diabatic descent cannot explain this behaviour, particularly when the corresponding profiles of N₂O are inspected in Fig. 55. These profiles show the effects of averaging of outside and inside air above θ ≈ 450 K during the second and fourth phases, but it is clear that descent of the phase 1 profile cannot explain the N₂O between 410 K and 450 K during phases 2–4. A consistent explanation is that the increase above 410 K in phases 3 and 4 was caused by evaporation of HNO₃-containing particles falling from above. Below θ ≈ 410 K, it seems that this mechanism enhanced F(NO₂) below 410 K in phase 2, but that by phases 3 and 4 transport of denitrified air from deep in the vortex toward the outer vortex caused the observed decreases (which extended up
to $\theta \approx 430$ K in phase 4). The slope of the vertical profile of $R(\text{H}_2\text{O})$ remained constant above $\theta \approx 400$ K for the first three phases (Fig. 56), with dehydration becoming evident below $\theta \approx 450$ K in phase 4, and below $\theta \approx 400$ K in phase 3. These results show again that denitrification and dehydration were separate processes, with the NO$_x$ presumably falling out first on smaller particles, which are more prone to evaporate before falling out of the stratosphere.

5. MID LATITUDES

(a) Horizontal structure

There is always structure in tracer mixing ratios during horizontal flight, down to the smallest scales resolvable by 1 Hz instruments operating on the ER2, which flies at 200 m s$^{-1}$. There is also small-scale structure in wind speed and direction, and in the isentropes mapped by the microwave temperature profiler. Figure 57 shows N$_2$O, wind speed, wind direction and the isentropes for 940603; in addition to the small-scale structure another, large-scale, feature is evident: along the near-isentropic flight track, there is a low-high-low structure in N$_2$O, which is crudely mirrored in the PV analysis at the time of the flight (Fig. 58), although not at the exact location of the flight track. This feature was sufficiently common that it is evident in medians from the AAOE (Fig. 59), and corresponds to air with low absolute PV being advected eastward and poleward, splitting-off high absolute PV air from the vortex edge which is displaced equatorward. Mid-latitude air is largely the resultant of peel-off air from the low absolute PV reservoir in the tropics interleaved with high absolute PV air from the vortex. It should be noted that the small-scale structure in the isentropes shown in Fig. 57(b) occurred over the Southern Ocean.
near the date-line, with no land upwind for several thousand kilometers; these variations, combined with the small-scale variations in wind speed and direction, will place a limit on how well the advection of tracers can be modelled; their kinematic effects are likely to stimulate mixing. The diabatic cooling rates shown above constitute a further limit to the ability of contour-advection techniques to represent structure in tracer fields.

We also reconsider here the latitudinal plots of \( F(\text{NO}_x) \) for the southern mid latitudes. In late March/early April, the horizontal gradient was equatorward, with the lowest values near 23°S and the highest values at high latitudes (Fig. 60). Combined with the scatterplots of \( \text{NO}_y \) versus CO and CNC number density in the tropics which were discussed earlier, the source of the low \( F(\text{NO}_x) \) in the southern hemisphere during phase 1 is, therefore, unlikely to have arisen as a memory of the denitrified 1993 vortex, which broke up in November of that year. There was an increase in \( F(\text{NO}_x) \) above Christchurch from March to October at \( \theta > 400 \) K (Fig. 61). However, the positive lapse rate did not clearly develop until phases 3 and 4; before that there was no clear lapse rate, and the values of \( F(\text{NO}_x) \) increased little.
Figure 58. 00 UTC 940604 450 K potential vorticity (PV) plots corresponding to Fig. 57. (a) UKMO operational analysis and (b) ECMWF analysis. Note the high–low–high structure in the absolute value of PV near, but not actually along, the flight track (shown by a black line), corresponding to the low–high–low structure in the N$_2$O. Neither of the analyses centered on the mid point of the flight (00 UTC 940604) accurately correlates with the N$_2$O structure however; this was true for all available analyses.
Figure 59. Medians of N$_2$O for all the AAOE flights, relative to wind speed maximum. Note the low--high--low pattern. Potential temperature $\theta = 425 \pm 10$ K.

Figure 60. Horizontal F(NO$_y$), means at potential temperature $\theta \geq 440$ K for phase 1 of the ASHOE from 25°S to 70°S.

Figure 61. Profiles of potential temperature vs. mean F(NO$_y$) near Christchurch, for each of the four phases of the ASHOE/MAESA.
Figure 62. Profiles of potential temperature vs. mean N₂O near Christchurch, for each of the four phases of the ASHOE/MAESA.

Figure 63. As Fig. 62 but for mean R(H₂O).

Figure 64. Mean profiles for the ASHOE flights in the upper troposphere and lower stratosphere, pressure altitude relative to the tropopause for (a) grand average of CFC-11 over all phases, and (b) the averages of CO for each of the four phases.
between phases 1 and 2. This pattern is not consistent with the pattern of gas-phase diabatic descent which was greatest between phases 1 and 2, as shown by the N₂O profiles in Fig. 62, but it is consistent with the effects of the evaporation of HNO₃-containing particles falling from above at the edge of the vortex followed by equatorward transport via peel-off. The variability shown in Fig. 61 below θ = 400 K during all phases shows that F(NO₃)there is determined by injection across the tropical tropopause of high but variable F(NO₃) air, mixed with air from the vortex. From midwinter on, the latter could be either denitrified or enhanced. The residual water profiles at Christchurch are essentially constant at 4.0 to 4.2 p.p.m.v. above θ = 400 K for the first two phases, but show an increase of a few tenths of a p.p.m.v. between 400 and 475 K between phases 2 and 3 (Fig. 63). Again, this is not explicable by diabatic descent in mid latitudes, and the N₂O actually decreased, so poleward transport of tropical air moistened by the warmer equatorial tropopause during northern summer is not a viable explanation either. Evaporation of ice crystals falling from above at the edge of the vortex is, however, a consistent explanation, and may account for much of the behaviour below 400 K as well, although near 350 K there may have been some evidence of equatorward transport of antarctic drying during phase 3.

The many flights into and out of Christchurch gave a set of vertical profiles there through the tropopause, and allow examination of mean tracer slopes either side of it. We consider two tracers, CFC-11 and CO: each has a decrease with height in the upper troposphere. The grand average is shown for CFC-11, since the samples are sparser and it was not measured on dynamics and radiation flights (Fig. 64). As in the tropics, there is a detectable ‘standing crop’ of recently stratospheric air in the upper troposphere over Christchurch. This feature was also visible in the smaller number of flights from Moffett Field. Figure 64(b) suggests that the signature of a fraction of recently tropical air extends higher than θ ≈ 400 K, to 450 K. The residual water profiles in Fig. 63 reinforce this conclusion. CO, with its shorter lifetime, is a more sensitive indicator than the longer-lived tracers, and the residual water also makes the mixing of recently tropical air out to 44°S at up to θ ≈ 450 K more apparent.

6. THE θ ≈ 400 K TRANSITION

The vertical profiles of many chemical species show a change of slope close to θ ≈ 400 K, from high through middle and subtropical latitudes to the inner tropics, in both hemispheres. The transition tends to be somewhat higher in the arctic winter, some 415 K, than in the antarctic (Tuck 1989; Proffitt et al. 1990, 1993; Tuck et al. 1992). In each polar case the transition is about a scale height or Δθ ≈ 100 K above the tropopause. We exemplify the transition here with grand average water profiles at 37°N, 21°N, 0° ± 7°, 18°S, 44°S and 67°S (Fig. 65(a)). It is interesting to note that the θ ≈ 400 K level is above the maximum equilibrium level which can be reached by surface air in the tropics. However, there is clear evidence from ²²⁳Rn observations simultaneously with total water and water vapour which showed penetrations of tropospheric air to θ ≈ 410 K (Kelly et al. 1993; Kritz et al. 1993) north of Australia in January and February 1987. Indeed, saturation with respect to ice was encountered briefly in horizontal flight at θ ≈ 410 K in the tropics southbound from Hawaii on 941026, and there was a substantial stretch of track close to it. The mean tropical tropopause is at about θ ≈ 360 K. Given the evidence for horizontal exchange with mid latitudes shown in section 3, which included advection of the tracer abundances and near isothermal temperature profiles characteristic of mid latitudes into the inner tropics, it is not surprising that the θ ≈ 400 K transition is found over the entire globe. It is also evident in PV analyses which are completely independent of the airborne tracer measurements (Tuck 1989). It seems likely that the transition layer
Figure 65. (a) Grand average profiles for the ASHOE/MAESA near 37°N, 21°N, 0° ± 7°, 18°S, 44°S and 67°S of potential temperature (θ) vs. water vapour. Note the transition to low stratospheric values at θ ≈ 400 K, with intermediate values between there and the tropopause (θ ≈ 360 K in the tropics, ≈ 330 K in mid latitudes, ≈ 300 K near Antarctica). The values near the equator show the effects of dehydration there, in contrast to the others. Also shown are mean total water profiles relative to the tropopause during phase 1, (b) near 37°N, and (c) near 44°S.

owes its existence to the synoptic-scale activity in the troposphere, and the need to absorb the upward propagation of such events in the lowest layer of the stratosphere (Charney and Drazin 1961). Certainly, in polar regions and mid latitudes the windfield and tracer patterns in this layer are the direct result of the evolution of tropospheric trough/ridge systems. The water content between the tropopause and 400 K in the extratropics (Figs. 65(b) and (c)) can only arise from injection of air from the extratropical troposphere.

The importance of the layer between the tropopause and 400 K may be two-fold in the consideration of problems connected with global change. Firstly, the maximal ozone loss occurring in it during polar winter, particularly over Antarctica, is more readily transmitted
to middle latitudes than that occurring above $\theta \approx 400$ K (Murphy 1991). Secondly, the water vapour content there is clearly a mixture of upper tropospheric air with the dry stratospheric air above, and is more variable. The moist air cannot have come from the tropics. Because water above the mid troposphere plays a prominent role in amplifying the increase in surface warming in model calculations of the atmospheric response to an increase in greenhouse gases, the content of this layer between the tropopause and $\theta \approx 400$ K could be important for climate. Values at the mid-latitude tropopause ranged up to 50 p.p.m.v. in the northern hemisphere, and to about 60% of this value in the southern hemisphere.

7. DISCUSSION AND CONCLUSIONS

The Brewer–Dobson circulation as originally formulated was based on advection in a meridional plane, with upward motion in the tropics (necessitated by the need to dry air at the tropical tropopause) and downward motion elsewhere. Since the predominant winds in the stratosphere are westerly in winter and easterly in summer outside the tropics, and alternate between these directions in a quasi-biennial manner in the tropics, it is apparent that the construct does not represent the physical, three-dimensional motion of the air; it is a statistical, long-term mean. If the construct were to be taken literally, tropospheric air would be found below, at and immediately above the tropical tropopause, with stratospheric air above, at and immediately below the extratropical tropopause. In fact there is a permanent, standing fraction of stratospheric air detected in the upper tropical troposphere; there is a similar standing fraction of tropospheric air detected above the extratropical tropopause, in the lower stratosphere. Recirculation of air between the upper troposphere and the lower stratosphere is thus an important concept. The presence of air in the upper tropical troposphere which had been in the extratropical lower stratosphere places, in particular, an important constraint on simplified concepts such as residence times, the age of the air, and a tropical ‘pipe’ for vertical transport. The evidence for transport into the tropics from mid latitudes showed it to be even more extensive above the tropopause; up to at least $\theta \approx 480$ K, a substantial fraction (1/4 to 3/5, approximately) was found to have recently come from mid latitudes. It was also observed that the near-isothermal profiles characteristic of the extratropical stratosphere sometimes occurred at and above the inner tropical tropopause, in association with dry, relatively low-tracer air. The advection of such thermal profiles into the equatorial region will allow deeper penetration of cumulonimbus tops into the stratosphere, compared with the temperature inversion otherwise found there. The minimum in total water (the hygropause) was observed to be sometimes, but not invariably, coincident with the tropopause over a wide range of latitudes (21°N to 24°S) from late March to late October at longitudes near the date-line. This is not consistent with the idea that entry of air to the stratosphere is confined to the Micronesia region during boreal winter; however, on only one flight in late October was anything resembling subvisible cirrus detected at or near the tropopause. Examination of aerosol observations reveals that the total number-density of small particles showed a maximum in the air near the tropical tropopause which had the minimum total water content, i.e. in dehydrated air. This suggests that the numerous small particles may be formed when ice crystals start to fall out, leaving still cold air but with no surfaces upon which the condensable vapours (HNO$_3$, H$_2$SO$_4$, H$_2$O) may deposit. Homogeneous nucleation is a possible mechanism.

A further interesting feature of the tropical flights was the asymmetry in the northern hemisphere and southern hemisphere observations of many of the trace species, particularly those with pronounced asymmetries in their tropospheric sources and abundances,
such as CO and total condensation nuclei. The asymmetry extended to the highest latitudes investigated, showing that tropical air mixes out sufficiently rapidly to maintain the tropical tropopause signatures there. Interestingly, NO\textsubscript{x} exhibited such behaviour during late March. Such asymmetries were observed at the tropical tropopause and above it up to θ ≈ 480 K; in mid latitudes the asymmetry extended to 10 km above the tropopause for CO. The conclusion to be drawn from this appears to be that much of the air in the northern hemisphere stratosphere originated in the northern hemisphere troposphere, with the southern hemisphere stratosphere being fed mainly from the southern hemisphere troposphere. This conclusion could be important from the point of view of chemical species whose stratospheric partitioning is particle-sensitive (such as nitrogen oxides or inorganic chlorine), since the northern stratosphere will have a greater particle loading than its southern counterpart. There is also the unexplored possibility that the aerosol loading in the northern stratosphere could be chemically different than in its southern counterpart. The greater NO\textsubscript{x} values at the tropical tropopause in the northern tropics compared with the southern tropics in March are of unknown origin. It could arise from greater industrial activity, but it could also be the result of a greater fraction of continental than maritime deep convection in the northern tropics. The recirculation of ‘older’ stratospheric air from mid latitude into the tropics is probably more vigorous in the northern hemisphere, particularly in winter; this mechanism too would be consistent with the observations. At near-tropospheric mixing ratios of N\textsubscript{2}O (roughly 300–310 p.p.b.v.), the width of the NO\textsubscript{x} distribution as a function of N\textsubscript{2}O was broadest in the northern tropics in late March, and in the southern tropics in late October. These broader NO\textsubscript{x} distributions at high N\textsubscript{2}O appeared to originate from high-NO\textsubscript{x} air (relative to N\textsubscript{2}O) in the northern hemisphere, but from low-NO\textsubscript{x} air (relative to N\textsubscript{2}O) in the southern hemisphere. In late March, the southern tropics contained low-NO\textsubscript{x} air (relative to N\textsubscript{2}O); the quantity F(NO\textsubscript{x}) increased polewards from 23°S to 68°S, suggesting a tropical source for such air.

The downward branch of the circulation in winter occurs largely in association with the polar-night jet/vortex system, where the diabatic descent is largest. This is evident both from the tracer distributions and from diabatic cooling rates calculated from high spectral resolution observations of the upwelling long-wavelength radiation, and has important implications for how high-latitude air is transported to middle latitudes: does it occur throughout the winter, or in essence only upon the spring break up of the vortex? Since ozone loss induced by halogen chemistry, modulated by the heterogeneous effects of PSCs, occurs in the winter stratospheric vortices, the question is of direct relevance to the observed loss of ozone in mid latitudes. If the view is taken that the downward branch of the diabatic circulation is driven by mechanically forced excursions of mid-latitude air to the polar regions where it will be too warm relative to the zonal mean radiative field and hence will cool, then the evidence for such excursions becomes important. However, mid-latitude air advected poleward in winter ascends the cold polar dome in the isentropes, and is cooled adiabatically, so that it may be, depending on the individual synoptic situation, colder rather than warmer than its environment on arrival at the vortex (Tuck 1989). The essential effect is that entropy should be lost, in order to support the downward motion.

It has been shown that, in 1987, operational meteorological analyses underestimated the extent of penetration of upper tropospheric/lower stratospheric ridges over Antarctica (Tuck 1994); the spectrally resolved infrared radiative observations in 1994 showed the presence of a tongue of warm air between about 330 and 450 K potential temperature in the outer vortex, which was poorly represented by both the TOVS operational satellite sounding system and the operational meteorological anlayses. This feature, combined with a careful handling of the emission in the water vapour rotational band from 15 to 100 μm wavelength (which emits a large fraction of the energy at the temperatures of the lower
Figure 66. \( N_2O \) medians, potential temperature \( \theta = 450 \pm 10 \text{ K} \), relative to the wind speed maximum. The five labelled curves are the AAOE in 1987 from South America and the four phases of the ASHOE/MAESA in 1994 from New Zealand. The values are lower and the gradients tighter for the AAOE data.

Stratospheric antarctic vortex), led to estimates of the diabatic cooling rates in the outer vortex of 1 to 3 K day\(^{-1} \) in the range 400 < \( \theta < 500 \text{ K} \) (15–20 km), and values from \( \sim 0.2 \text{ K day}^{-1} \) at the tropopause (\( \theta \approx 300 \text{ K} \)) to \( \sim 1 \text{ K day}^{-1} \) at \( \theta \approx 400 \text{ K} \). The tongue of warm air will obviously cool by net emission of radiation to the cold air above and below it as well as to space. These values are about a factor of 2–3 larger than those reported for free-running model calculations, and imply a larger flow of air through the vortex than such calculations. Such cooling rates also imply that a larger amount of mixing across the jet can be sustained, and indeed evidence of such mixing was found in the form of meridional gradients of tracers across the jet core. The 10° latitude-wide belt centred on the jet maximum was shown to be a mixing zone; the mass in the belt centred at 65°S is larger than that in the cap poleward of it, so the maintenance of the observed radial gradients implies substantial mixing in the outer vortex across the jet core. Individual flights showed multiple edges in the quasi-isentropic tracer distributions, supporting the notion of exchange. It was also shown that the condensable vapours, \( \text{NO}_x \) and \( \text{H}_2\text{O} \) showed evidence in the outer vortex of evaporation of particles falling from above, making the use of denitrified and dehydrated air from the antarctic vortex as markers of the transport of vortex air to mid latitudes more problematical than hitherto appreciated. Both inside and outside the vortex, the air encountered in the New Zealand sector in 1994 was wetter than that observed in the South American sector in 1987. Single-aircraft data cannot unequivocally resolve the question as to whether this is a longitudinal or a temporal effect, but evidence for both factors was found. The air at middle and high latitudes of both hemispheres in 1994 was drier, in the sense of having less total hydrogen, than the air encountered in the tropics, and was about 3–4 years ‘older’ on average, indicating a stratospheric moistening trend over that period. There is a difference in the tracer gradients between the AAOE in 1987 and the ASHOE/MAESA in 1994, see Fig. 66. The gradients at the same potential temperatures (\( \theta = 450 \pm 10 \text{ K} \)) are slacker in the New Zealand sector across the jet core, and the values are higher than they were in the South American sector. This is consistent with a picture in which dry vortex air is produced over West Antarctica, and tends to be peeled off between the peninsula and Greenwich meridian, with moister mid-latitude air mixing into the outer vortex over East Antarctica. Note also that the low–high–low \( \text{N}_2\text{O} \) pattern is evident in these medians, although to differing degrees at different times. The above points tend to argue that the water differences between the missions are partly the result of a trend in time, and are partly a longitudinal effect. However, it will take
satellite observations to establish this conclusively, particularly as regards the longitudinal gradients. The maintenance of a longitudinal gradient at latitudes 40°S–60°S in water by dehydration processes occurring inside the vortex would support the idea that there is substantial flow through the system.

The air in mid latitudes shows filamentary structures on all meridional scales from 200 m to several hundred kilometres, which may be traced back to both tropical and high-latitude sources. Contour-advection studies (Pierce and Fairlie 1993; Waugh et al. 1994) suggest that the zonal scale of some of these filaments may be thousands of kilometres. It was apparent that in the approximately one-scale-height-deep layer between the tropopause and θ ≈ 400 K, there was efficient exchange with both the polar regions and the tropics. The ozone and tracer content of this layer is the result of ‘old’ air from polar descent being mixed with some ‘young’ air from the tropics plus further contribution from the mid-latitude troposphere. The convolution of its composition by numerical models demonstrates the presence of a standing crop of identifiable stratospheric air.

For the aircraft-accessible domain (up to θ ≈ 530 K) the tracers show sharp horizontal gradients between the inner tropics and mid latitudes, and between mid latitudes and the vortex. In the vertical there are sharp gradients at the tropopause and a less sharp transition at θ ≈ 400 K over the entire globe. Examination shows that these gradients are maintained by processes of fluid mechanical stripping of filaments or sheets of air by differential advection (wind shear), which also involve exchange of air across the ‘boundaries’; demonstration of this is to be found by a mean detectable presence of trans-boundary air and mean mixing slopes. The convolution of the material surfaces suggests that fluxes across these gradients may be difficult to calculate; it is possible that a more reliable approach would be to use the observed tracer distributions as a test of how well model calculations are representing the interplay between the radiative and fluid mechanical processes.

The interpretation of the observations offered here suggests that forcing by tropospheric meteorological processes—deep convection in the tropics and synoptic troughridge systems in the extratropics—is central to understanding the lower stratosphere up to about 21 km altitude. Although the aircraft data do not include observations of the summer hemisphere, and obviously say nothing directly about the middle and upper stratosphere, they do make it clear that a simplified construct of advection in two dimensions (altitude and latitude) is inadequate to understand the maintenance of the composition of the lower stratosphere. As regards ‘forcing’ and ‘control’ (see, for example, Holton et al. 1995), such ideas need to be used with caution in a nonlinear system such as the atmosphere with oceans and surface topography. It is manifestly obvious, however, that in the case of the lower stratosphere the seasonal cycle arising from the passage of the sun through the global sky causes the formation of the winter stratospheric vortex, and that the annual seasonal cycles in the northern and southern tropospheres affect both the way tropospheric air enters the stratosphere in the tropics, and exchanges across the vortex, sub-tropics and tropopause elsewhere. If there is control of the lower stratosphere it is by synoptic-scale systems in the troposphere. If there is a conceptual need to think of cause and effect, then it is clear that the atmosphere/surface system is driven by a radiative energy excess at low latitudes and a deficit at high latitudes (Simpson 1929; Ludlam 1980), and responds by deep convection in the tropics, whose occurrence is modulated by upper tropospheric penetrations of extratropical air as the result of tropospheric cyclogenesis in mid latitudes.

An alternative is to consider the entropy balance (O’Brien and Stephens 1995). The solar beam of high-energy photons incident upon the earth is a low-entropy state, while
the outgoing flux over $4\pi$ solid angle of low-energy terrestrial photons is associated with high entropy ($dq/T$ at $\sim 5800$ K versus $dq/T$ at $\sim 245$ K). The fraction of the entropy production associated with the atmosphere occurs mainly in the low-latitude troposphere, not the extratropical mid stratosphere. Even in the mid stratosphere, the absorption of the solar beam of ultraviolet and visible photons at low latitudes to produce oxygen atoms and ozone, whose translational energy is absorbed by the thermal bath of $O_2$ and $N_2$ molecules, produces entropy, and hence upward motion. This process would occur in the absence of Rossby waves. At middle and high latitudes, particularly in winter, air must lose entropy to descend; it does so by radiation to space, most efficiently in air parcels mechanically forced to move poleward from lower latitudes. An interesting question as regards the role of Rossby waves is whether or not they are that realization of the adiabatic dynamical response to the diabatic forcing from the sun which maximizes the entropy production.

In the context of longer time-scales one can also recall what is known of the history of the atmosphere–earth–sun system in considering the idea of control. Over geological time, the planetary rotation rate has decreased substantially, the continents have drifted and the solar output is thought to have increased. It is unlikely that Rossby-wave activity in the mid stratosphere remained unaffected. Of the three radiatively active atmospheric constituents, the well-mixed one, carbon dioxide, is known to have fluctuated over geological time in response to the temperature-dependent sea water and weathering chemistry of carbonates, and ozone must have, given that the present oxygen abundance dates from ‘only’ about 600 million years ago. Indeed, the chemical composition of the atmosphere is greatly affected by the presence of life, being many orders of magnitude removed from thermochemical equilibrium (Lewis and Randall 1923; Lovelock and Margulis 1974); the equilibrium state would be a solution of 0.1M nitric acid, $\text{HNO}_3$, in water rather than our $\text{N}_2–\text{O}_2$ atmosphere. The most constant feature appears to be the fact that the oceans have never frozen; there is a case for arguing that water exerts control in the long term. In the short term, the troposphere is convectively unstable because of the cooling to space by water vapour in the upper troposphere (Ludlam 1980); radiative equilibrium is not possible with liquid water at the surface, which must evaporate because the equilibrium vapour pressure above it produces a water vapour column in the lower troposphere which is optically thick enough to maintain surface temperatures above freezing. The flux of precipitation to the surface and transport of heat and water to high latitudes prevents runaway. Finally, we note that Johnson (1989) has convincingly argued, using isentropic analysis, that global monsoonal circulations are the response to the global-scale differential thermal forcing, pressure torques and frictional forces at the surface.

We may consider the lower stratosphere in practical terms: that of the injection of $\text{NO}_x$ and particles by aircraft. We have seen that the details of the transport upward across the tropical tropopause matters, as witnessed by the interhemispheric asymmetries in the lower stratosphere in these quantities and in CO. The ‘standing crop’ of identifiably stratospheric air in the upper troposphere is found at all latitudes. It would be difficult to deduce the presence of these signatures, let alone quantify them, by means of a radiative calculation on the $\theta \approx 400$ K surface. The same is true of the upper tropospheric air injected into the layer between $\theta \approx 400$ K and the tropopause in mid latitudes. There is no obvious reason why either the mass or the chemical content of the layer between 400 K and the tropopause should be constant; both the rate at which air enters it from below in the tropics and from above at high latitudes is variable and determined by the underlying tropospheric synoptic evolution. In the particular case of $\text{NO}_x$, we have seen that lightning in the tropics and gravitational settling and subsequent evaporation of PSC particles play a role, and that there is substantial lateral exchange of air between the tropics and the remainder of the atmosphere, up to at least $\theta \approx 480$ K and probably up to 550–600 K.
We conclude by noting the following results, all of which apply strictly only to the longitude sector between 120°W and 150°E:

(a) The evidence for recirculation of air between the upper troposphere and lower stratosphere, particularly in the tropics. Some air in the upper troposphere and lower stratosphere of the tropics contained depleted layers of tracers and near-isothermal temperature profiles characteristic of the mid-latitude lower stratosphere. Such advection would allow deeper convection than the temperature inversion otherwise found in the lower tropical stratosphere.

(b) The possibility that new particles form near the tropical tropopause as a consequence of the dehydration process.

(c) Interhemispheric asymmetries in CO, condensation nuclei and NO\textsubscript{y}, which suggest that the stratosphere in each hemisphere receives a dominant fraction of its air from its own troposphere. The Inter Tropical Convergence Zone is thus not a completely efficient mixer of tropospheric air from the two hemispheres.

(d) From the inner tropical tropopause up to $\theta \approx 500$ K there is an extensive fraction (1/4–3/5) of air recently from the lower stratosphere of mid latitudes.

(e) The layer between the tropopause and $\theta \approx 400$ K consists of air descended through the polar vortex mixed with air recently from the upper extratropical troposphere and from the lower tropical stratosphere.

(f) There was a difference of $\sim 0.5$ p.p.m.v. in the mixing ratios of $2\text{CH}_4 + \text{H}_2\text{O}$ at ER2 cruise altitudes ($\theta \approx 470$ K) between the younger air in the tropics and the older air elsewhere, in both hemispheres in both March and October. The $\text{H}_2$ was essentially flat at $\sim 0.5$ p.p.m.v. These observations suggest a positive trend in stratospheric water of $\sim 0.15$ p.p.m.v. year$^{-1}$ for the period 1990–94. We note that this period coincided with the most protracted El Niño on record.

(g) The diabatic cooling rates $d\theta/dt$ ranging from 1–3 K day$^{-1}$ between $\theta = 350$ K and 500 K in the cold air of the winter/spring antarctic vortex; they are associated with the presence of a tongue of relatively warm air in the lower levels of the outer stratospheric vortex, combined with emission in the water vapour rotational bands from 15 to 100 $\mu$m wavelength in the far infrared.

(h) Redistribution of stratospheric NO\textsubscript{y} and H\textsubscript{2}O by evaporation of particles falling from aloft is evident near the antarctic polar-night jet stream. This phenomenon will hamper the use of dehydration and denitrification as markers for the spread of vortex air to mid latitudes.

(i) The transport of PSC-processed air with low tracer mixing ratios to mid latitudes was very evident in early June, a period when minimum vortex temperatures had dropped below 195 K but when the polar-night jet had attained only about two thirds its eventual strength.

(j) There was ozone loss in the outer vortex of up to 25% which had developed between early June and early August. Low-ozone air was observed outside the vortex in mid October.

(k) Meridional gradients of tracers such as CFC-11, methane and nitrous oxide, NO\textsubscript{y}, H\textsubscript{2}O and HCl suggest that there is a 10° latitude-wide mixing zone centred on the antarctic jet core between 16 and 20 km altitude; it contains about as much mass as the cap poleward of it. The effects of such mixing were evident out to about 40°S in the South America sector in 1987, and out to about 50°S in the New Zealand sector in 1994.

(l) Vertical profiles in the outer vortex of tracers, wind speed and wind direction show layered structures both at the edge of and inside the vortex which are almost ubiqui-
tous over the Southern Ocean thousands of kilometers from land. They are likely to cause irreversible mixing across the polar-night jet stream.

(m) There may be a longitudinal asymmetry in the water vapour distribution both inside and outside the antarctic vortex in late winter and early spring, with low values and tight horizontal gradients over West Antarctica, and high values with slacker horizontal gradients over East Antarctica. Analysis of satellite observations is required to establish whether or not this is true.

There are some possibly significant implications of these results. The presence of detectably stratospheric air in the upper troposphere, including the tropics, raises the possibility that the chemical production from the stratosphere, including its particle loading, could be involved in cirrus cloud formation in the upper troposphere and hence in climate. The diabatic cooling rates in the antarctic vortex are capable of sustaining a larger mass flow through it than has been possible in free-running model calculations, with unknown implications for the lifetimes of CFCs and $\text{N}_2\text{O}$. The lower stratospheric asymmetry in particle number-density between the hemispheres may have important implications for the chemistry associated with CFCs and high-flying aircraft, particularly if the asymmetry extends to aerosol composition as well as number. The combined radiative effects of ozone loss between the tropopause and $\theta \approx 400$ K, with the presence there of water vapour of mid-latitude tropospheric origin, raises the question of the stability of the tropopause under CFC and greenhouse forcing. The water vapour ranges from 5 to 50 p.p.m.v. between 400 K and the extratropical tropopause; the values at the tropopause are variable and in the high end of the range, and are radiatively significant, particularly at cold tropopausas.

ACKNOWLEDGEMENTS

The late Edwin F. Danielsen initially had the vision to see the value of and implement fast response in situ observations of trace species on the ER2. Thanks are due to the large team of scientists, engineers, aircrew and groundcrew from several countries who made the ASHOE/MAESA mission work. The cooperative attitude of the national meteorological services of the United States, United Kingdom, Australia, New Zealand and of the ECMWF is greatly appreciated. The Navy and the National Science Foundation of the United States provided facilities at Christchurch Airport.

REFERENCES


Ellsaesser, H. W.


Fahey, D. W., Kelly, K. K., Ferry, G. V., Poole, L. R., Wilson, J. C., Murphy, D. M., Loewenstein, M. and Chan, K. R.

1989 *In situ* measurements of total reactive nitrogen, total water and aerosol in a polar stratospheric cloud in Antarctica. *J. Geophys. Res.*, 94, 11299–11316

Fahey, D. W., Kelly, K. K., Kawa, S. R., Tuck, A. F., Loewenstein, M., Chan, K. R. and Heidt, L. E.

1990 Observations of denitrification and dehydration in the winter polar stratospheres. *Nature*, 344, 321–324

Farman, J. C., Gardiner, B. C. and Shanklin, J. D.

1985 Large losses of total ozone in Antarctica reveal seasonal NO<sub>x</sub>/ClO<sub>x</sub> interaction. *Nature*, 315, 207–210


Ferry, G. V., Neish, E., Schultz, M. and Pueschel, R. F.

1964 ‘Photochemical behaviour of the ozone layer’. Canadian Armaments Research and Development Establishment, Technical Note 1627

Hampson, J.


Hanson, D. R. and Ravishankara, A. R.

1991 The reaction probabilities of ClONO<sub>2</sub> and N<sub>2</sub>O<sub>5</sub> on 40 to 75% sulfuric acid solutions. *J. Geophys. Res.*, 96, 17307–17314

Harries, J. E.

1994 Reactive uptake of ClONO<sub>2</sub> onto sulfuric acid due to reaction with HCl and H<sub>2</sub>O. *J. Phys. Chem.*, 98, 5728–5735

Harrison, H.


Harries, J. E.


Johnson, D. R.  
Johnston, H. S.  
Juckes, M. N. and McIntyre, M. E.  
Kawa, S. R., Fahey, D. W., Kelly, K. K., Dye, J. E., Baumgardner, D., Gandrud, B. W., Loewenstein, M., Ferry, G. V. and Chan, K. R.  
Kelly, K. K., Tuck, A. F., Heidt, L. E., Loewenstein, M., Podolske, J. R., Strahan, S. E. and Vedder, J. F.  
Kelly, K. K., Proffitt, M. H., Chari, K. R., Loewenstein, M., Podolske, J. R., Strahan, S. E., Wilson, J. C. and Kley, D.  
Kriz, M. A., Rosner, S. W., Kelly, K. K., Loewenstein, M. and Chan, K. R.  
Lewis, G. N. and Randall, M.  
Loewenstein, M., Podolske, J. R., Chan, K. R. and Strahan, S. E.  
Lovelock, J. E. and Margulis, L.  
Ludlam, F. H.  

1987 A high resolution one-layer model of breaking planetary waves in the stratosphere. *Nature*, 328, 590–596  
1979 *In situ* measurements of the mixing ratio of water vapor in the stratosphere. *J. Atmos. Sci.*, 36, 2513–2534  
1974 Atmospheric homeostasis by and for the biosphere: the gaia hypothesis. *Tellus*, 26, 2–10  
1980 *Clouds and storms*. Chapter 1.2. Pennsylvania State University Press, College Park, USA


NASA 1994 'Airborne Southern Hemisphere Ozone Experiment/Measurements for Assessing the Effects of Stratospheric Aircraft'. Project Office, NASA Ames Research Center, Moffett Field, California, USA


Reed, R. J. and German, K. E. 1965 A contribution to the problem of stratospheric diffusion by large-scale mixing. Mon. Weather Rev., 93, 313–321


