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ATMOSPHERIC TRACERS
- A REVIEW -

BY

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ATMOSPHERIC TRACERS - A REVIEW

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ABSTRACT

A brief presentation of the various basic equations specifying physical processes in the atmosphere suggests practical limitations in elucidating these processes in various time scales and regions of the atmosphere.

The conservatism of various tracers and their measurement is discussed. The tracers considered most appropriate to various spatio temporal scales of atmospheric system are collated and presented, and finally discussed in relation to mass transfer models within the stratosphere and through the tropopause.

TRACEURS ATMOSPHÉRIQUES - REVUE

par

A. D. Christie

RÉSUMÉ

Dans un bref exposé des diverses équations fondamentales spécifiant les processus physiques dans l'atmosphère, l'auteur propose des limitations pratiques pour élucider ces processus dans diverses échelles de temps et différentes régions de l'atmosphère.

L'auteur traite du conservatisme des divers traceurs et de leur mesure. Il collationne et présente les traceurs considérés comme les plus appropriés aux diverses échelles spatio-temporelles du système atmosphérique et en établit ensuite la relation avec les modèles de transfert de masse dans la stratosphère et à travers la tropopause.

ATMOSPHERIC TRACERS - A REVIEW

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A. D. Christie

1. Introduction

Certain physical properties of a fluid and trace constituents of the atmosphere may be conservative under limiting conditions discussed later, and may be used to infer information on transfers. The relevant continuity equations will be presented and their usefulness illustrated in interpreting various tracer distributions.

2. The Equations specifying Atmospheric Transfers

We will now present the various smoothed equations specifying fluid flow in the atmosphere, and see what their implications are in investigating transfer.

In all subsequent sections the following notation will be used:

ρ : density

α : specific volume

P : pressure

$\underline{V}$: velocity

u : zonal component of velocity

v : meridional component of velocity

ω : vertical component of velocity

s : any conservative entity in amount per unit mass of air

$\underline{\Omega}$: angular velocity of the earth's rotation

ψ : potential energy

T : Temperature

θ : potential temperature

C_p : specific heat at constant pressure

R : gas contant for 1 gm. dry air

q : energy added to unit mass of system by diabatic heating

λ : latitude

ϕ : longitude

η : absolute vorticity

$\underline{P}$: potential vorticity

$\underline{F}$: frictional force per unit volume

τ_x : $\underline{i}\,\tau_{xx} + j\,\tau_{xy} + \underline{k}\,\tau_{xz}$ is the x component of the viscous stress, tensor, where $\underline{i}$, $\underline{j}$, $\underline{k}$ are unit vectors in the x, y and z directions respectively.

Vector quantities are denoted by means of a sub-bar, e.g. $\underline{V}$. A mean value is defined by an integral of the form $\bar{p} = \frac{1}{\tau}\int_{t-\tau/2}^{t+\tau/2} p\, dt$, and at any instant, the field may be represented by two components, the mean and the deviation as follows: $p = \bar{p} + p'$, where p' is the instantaneous deviation from the mean value.

In the atmosphere $\frac{\rho'}{\rho} \doteq 10^{-3}$ whereas $\frac{\underline{V}'}{\underline{V}} \doteq 1$ which allows the approximation $\rho \doteq \bar{\rho}$ in most instances.

The equation of continuity of mass is:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \rho\, \underline{V} = 0$$

By introducing the notation of means and deviations, then temporally averaging, we obtain:

$$\frac{d\bar{\rho}}{dt} = -\bar{\rho}\nabla \cdot \overline{\underline{V}} \qquad 2.1.$$

If we postulate the existence of any conservative entity, s, per unit mass of air:

$$\frac{\partial \rho s}{\partial t} = - \nabla \cdot \rho s \underline{V}$$

$$\text{i.e.} \quad \frac{\partial \bar{\rho}(\bar{s} + s')}{\partial t} = - \nabla \cdot \bar{\rho}(\bar{s} + s')(\overline{\underline{V}} + \underline{V}')$$

Taking means of both sides and subtracting 2.1.

$$\bar{\rho}\frac{d\bar{s}}{dt} = - \nabla \cdot (\bar{\rho}\,\overline{s' V'}) \qquad 2.2.$$

The equation of motion for unit mass of air derived from Newton's law of rate of change of momentum, in terms of velocities measured relative to a system of axes rotating with the earth, may be expressed as follows:

$$\frac{d\overline{V}}{dt} = -2\underline{\Omega} \times \overline{\underline{V}} - \nabla\psi - \bar{\alpha}\nabla\bar{p} + \bar{\alpha}\nabla \cdot \begin{array}{c|ccc} & i & j & k \\ \hline i & \tau_{xx} - \bar{\rho}\,\overline{u'u'}, & \tau_{xy} - \bar{\rho}\,\overline{u'v'}, & \tau_{xz} - \bar{\rho}\,\overline{u'\omega'} \\ j & \tau_{xy} - \bar{\rho}\,\overline{u'v'}, & \tau_{yy} - \bar{\rho}\,\overline{v'v'}, & \tau_{yz} - \bar{\rho}\,\overline{v'\omega'} \\ k & \tau_{xz} - \bar{\rho}\,\overline{u'\omega'}, & \tau_{yz} - \bar{\rho}\,\overline{v'\omega'} & \tau_{zz} - \bar{\rho}\,\overline{\omega'\omega'} \end{array}$$

where the forces acting on the unit mass are, in order of appearance on the righthand side of this equation.

1. The coriolis force resulting from the rotation of the system.

2. The geopotential resulting from the net component of gravity and centrifugal force.

3. The pressure gradient force.

4. The frictional force $\underline{F}$ resulting from the divergence of the viscous stresses τ_{ij} and the virtual eddy stresses $\bar{\rho}\,\overline{u'v'}$ etc.

This equation may be converted into the absolute vorticity equation by taking the curl of both sides.

$$\frac{d\underline{\eta}}{dt} = -\underline{\eta}(\nabla \cdot \underline{V}) + (\underline{\eta} \cdot \nabla)\underline{V} - \nabla\bar{\alpha} \times \nabla\bar{p} + \nabla \times \bar{\alpha}\,\underline{F} \qquad 2.4.$$

Since the atmosphere is not auto barotropic, i.e. $\nabla\bar{\alpha} \times \nabla\bar{p} \neq 0$ for all t, we will consider an alternative function, the potential vorticity which removes this term from the equation. By first adding the smoothed continuity equation $\frac{d\bar{\alpha}}{dt} = \bar{\alpha}\nabla \cdot \underline{V}$ to 2.4 and then multiplying by $\bar{\alpha} \cdot \nabla\bar{\theta}$, the equation becomes:

$$\frac{d}{dt}\left\{\bar{\alpha}\nabla\bar{\theta}\,\underline{\eta}\right\} = \bar{\alpha}\,\underline{\eta} \cdot \nabla\left(\frac{d\theta}{dt}\right) - \bar{\alpha}\nabla\bar{\theta} \cdot \nabla\bar{\alpha} \times \nabla\bar{p} + \bar{\alpha}\nabla\bar{\theta} \cdot \nabla \times \bar{\alpha}\,\underline{F} \qquad 2.5.$$

Furthermore, the second term on the right is identically zero since θ is a function of α and p.

Finally, the thermal transfer equation may be derived by subtracting the dynamic energy equation, from the first law of thermodynamics,

$$\frac{dT}{dt} = \frac{1}{C_p}\left[\alpha\frac{dp}{dt} + \frac{dq}{dt} + \alpha\left\{\underline{\tau}_x \cdot \frac{\partial \underline{V}}{\partial x} + \underline{\tau}_y \cdot \frac{\partial \underline{V}}{\partial y} + \underline{\tau}_z \cdot \frac{\partial \underline{V}}{\partial z}\right\}\right]$$

where $\frac{dq}{dt}$ is the rate of diabatic heating per unit mass of air.

Introducing the potential temperature enables us effectively to combine the individual rate of change of temperature and the adiabatic temperature change resulting from pressure variations as follows:

$$\frac{d\theta}{dt} = \frac{1}{C_p}\left(\frac{p_o}{p}\right)^K\left[\frac{dq}{dt} + \alpha\left\{\underline{\tau}_x \cdot \frac{\partial \underline{V}}{\partial x} + \underline{\tau}_y \cdot \frac{\partial \underline{V}}{\partial y} + \underline{\tau}_z \cdot \frac{\partial \underline{V}}{\partial z}\right\}\right] \qquad 2.6.$$

Smoothing, this becomes:

$$\frac{d\bar{\theta}}{dt} = \frac{1}{C_p}\left(\frac{P_o}{p}\right)^K \left[\frac{d\bar{q}}{dt} + \overline{\underline{V}' \cdot \nabla q'} + \alpha\left\{\underline{\tau}_x \cdot \frac{\partial \underline{V}}{\partial x} + \underline{\tau}_y \cdot \frac{\partial \underline{V}}{\partial y} + \underline{\tau}_z \cdot \frac{\partial \underline{V}}{\partial z}\right.\right] \qquad 2.7.$$

Clearly none of the properties whose total derivatives are evaluated in the above equations in the smoothed system may be considered conservative, unless the terms appearing on the right of the equations total zero.

3. Considerations of Tracer Conservatism in Practical Terms

3.1. Physical tracers

Various non-uniformly distributed physical properties of the atmosphere are shown in table 3.1 with a note on regions wherein conservatism may not be a plausible assumption. Sources and sinks are defined as regions in which the non-conservative effects result in positive and negative increments respectively to the mean value for that region.

3.1.1. Potential temperature, θ, and wet bulb potential temperature, θω.

$$\frac{\partial \theta}{\partial t} = -\underline{V} \cdot \nabla \theta + \frac{1}{C_p}\left(\frac{p_o}{p}\right)^K \left\{\frac{dq}{dt} + \alpha\left\{\underline{\tau}_x \cdot \frac{\partial \underline{V}}{\partial x} + \underline{\tau}_y \frac{\partial \underline{V}}{\partial y} + \underline{\tau}_z \cdot \frac{\partial \underline{V}}{\partial z}\right\}\right\} \qquad 2.6.$$

The potential temperature in the free atmosphere is of the order of 300°K and ranges over ten or more degrees at any level in a short period disturbance (36-48 hr). This rate of change is large compared with the average rates of heating or cooling (evaporation and condensation apart) of 1-2°K day^{-1}, but the latter is cumulative so θ becomes increasingly non-conservative with time.

Möller (1941), Staley (1958) and Manabe and Möller (1962) have calculated profiles of radiative cooling by water vapour for characteristic temperature and humidity distributions and show the radiative flux (which tends to destroy the curvature

TABLE 3.1

PHYSICAL TRACERS AND THEIR DEPARTURES FROM CONSERVATISM

Tracer	Source	Source Type	Sink	Sink Type
Potential Temp. θ	Atmosphere Earth	1. Radiation 2. Condensation 3. Frictional Dissipation	Atmosphere	1. Radiation 2. Evaporation
Wet Bulb Potential Temperature θ_{ω}	As above but condensation-evaporation sources and sinks now incorporated in the tracer function			
Absolute Vorticity	Atmosphere	Pressure-density solenoid field	Earth	Frictional Dissipation
Potential Vorticity	Atmosphere	Pressure-density solenoid field	Earth	Frictional Dissipation

of the lapse rate profile) is not seriously altered by large thermal and gaseous absorber gradients. If, however, the upper limit of haze or cloud layers corresponds to the lapse change, the radiative cooling may increase by orders of magnitude to a degree or more per hour, swamping other effects. Flux observations by Kuhn, Suomi and Darkow (1959) and Bushnell and Suomi (1961) show such excessive rates of cooling at cloud tops in mid-troposphere, suggesting it may not always be justifiable to assume conservatism for θ and θ_ω near cloud tops.

3.1.2. Absolute vorticity

The equation for absolute vorticity η is:

$$\frac{d\eta}{dt} = -\eta(\nabla \cdot \underline{V}) + (\eta \cdot \nabla)\underline{V} - \nabla\bar{\alpha} \times \nabla p + \nabla \times \bar{\alpha}\,\underline{F} \qquad 2.4.$$

so that η is conservative if:

1. the motion is non-divergent (incompressible fluid)
2. the tilting terms are zero (no deformation of the vorticity field)
3. the fluid has no (α, p) solenoids (barotropic) and
4. the motion be frictionless or $\bar{\alpha}\,\underline{F}$ irrotational

The equation for the vertical component (ζ + f) of the absolute vorticity η is:

$$\frac{d(\zeta + f)}{dt} \doteq (\zeta + f)\nabla_h \cdot \underline{V}_h + \left(\frac{\partial V_h}{\partial z} \times \nabla\omega\right) \cdot \underline{k} + \frac{\partial \omega}{\partial y} 2\Omega \cos\phi - \frac{\omega f}{r_e} \nabla_h \alpha \times \nabla_h p + \underline{k} \cdot \nabla \alpha\,\underline{F} \qquad 3.1.$$

The partial success of C. A. V. trajectories and barotropic forecasts suggest that for short periods the vorticity may be conserved in mid-troposphere ($\sim$ level of nondivergence) in the absence of baroclinic development. In the vicinity of the jet and in frontal zones in upper and lower troposphere the first, second and fifth terms on the right of 3.1 are each of magnitude 10^{-9} to 10^{-10} sec^{-1} and would be capable of producing large departures from conservatism ($\zeta + f \simeq 10^{-4}$ sec^{-1}) in a period of 12 hours.

3.1.3. Potential vorticity

The potential vorticity obeys the equation

$$\frac{d\overline{P}}{dt} = \frac{d}{dt}(\overline{\alpha}\nabla\overline{\theta}\cdot\underline{\eta}) = \overline{\alpha}\,\underline{\eta}\cdot\nabla\left(\frac{d\overline{\theta}}{dt}\right) + \overline{\alpha}\nabla\overline{\theta}\cdot\nabla\times\overline{\alpha}\,\underline{F} \qquad 2.5.$$

It is only conservative if the motion is adiabatic and either the fluid frictionless, the frictional force irrotational, or the gradient of potential temperature perpendicular to the rotation of $\overline{\alpha}\,\underline{F}$.

If $\underline{n}$ is a unit vector in the direction $\nabla\overline{\theta}$, the equation may be written:

$$\frac{d}{dt}\left(\overline{\alpha}\frac{\partial\overline{\theta}}{\partial n}\eta_n\right) = \overline{\alpha}\,\underline{\eta}\cdot\nabla\left(\frac{d\overline{\theta}}{dt}\right) + \overline{\alpha}\frac{\partial\overline{\theta}}{\partial n}R_n \qquad 3.2$$

where η_n is the component of absolute vorticity normal to the isentropic surface and $R_n = \underline{n}\cdot\nabla\times\underline{F} \doteq \left(\frac{\partial F_y}{\partial x} - \frac{\partial F_x}{\partial y}\right)_\theta$ denotes the component normal to the isentropic surface of the rotation of the frictional force.

The component of absolute vorticity η_n may be written to a close approximation as $\eta_n \doteq \zeta_n + f \doteq \zeta_\theta + f$ where ζ_n is the component of relative vorticity normal to the isentropic surface and ζ_θ is the relative vorticity evaluated from the projections on the horizontal of the horizontal wind components on the isentropic surfaces. Now writing $\frac{\partial}{\partial n} \doteq \frac{\partial}{\partial z} = -g\rho\frac{\partial}{\partial p}$ in equation: 3.2

$$\frac{d}{dt}\left[\frac{\partial\bar{\theta}}{\partial p}(\zeta_\theta + f)\right] = -\frac{\bar{\alpha}}{g}\,\eta\cdot\nabla\left(\frac{d\theta}{dt}\right)\ \frac{\partial\theta}{\partial p}R_n$$

$$\text{or}\ \frac{d}{dt}\left[-\frac{\partial\bar{\theta}}{\partial p}(\zeta_\theta + f)\right] = \frac{\bar{\alpha}}{g}\left\{\eta_z\frac{\partial}{\partial z}\left(\frac{d\theta}{dt}\right) + \eta_h\cdot\nabla_h\left(\frac{d\theta}{dt}\right)\right\} - \frac{\partial\theta}{\partial p}R_n \qquad 3.3$$

Staley (1960) has estimated the orders of magnitude of the terms on the right for large scale diabatic heating and friction to be as follows:

$$\frac{\bar{\alpha}}{g}\,\eta_z\frac{\partial}{\partial z}\left(\frac{d\theta}{dt}\right) \doteq 10^{-14}\,{}^{\circ}K\ \text{cm gm}^{-1}$$

$$\frac{\bar{\alpha}}{g}\,\eta_h\cdot\nabla_h\left(\frac{d\theta}{dt}\right) \doteq 10^{-15}\,{}^{\circ}K\ \text{cm gm}^{-1}$$

$$\frac{\partial\theta}{\partial p}R_n \doteq 10^{-15} - 10^{-16}\,{}^{\circ}K\cdot\text{cm gm}^{-1}$$

Since a representative value of potential vorticity in the lower stratosphere or frontal zone is 2×10^{-8} deg. gm.$^{-1}$ cm. sec., over a 12-hour period the largest non-conservative effect above would result in a change of at least an order of magnitude less than this value, implying that potential vorticity might be assumed conservative for periods of up to a day. Staley (1960) estimated the individual potential vorticity change by means of isentropic trajectories for an extratropical disturbance and inferred that in the vicinity of the frontal zone potential vorticity was not conserved. In fact, large positive potential vorticity changes occurred in the lower stratosphere and in the upper troposphere on the cold side of the front while large negative values occurred in the frontal zone and around the entire periphery of the cold trough in the upper troposphere.

Vorticity measurements necessitate an accurate description of the wind field. The network of meteorological observing stations is sparse and the observational accuracy diminishes with height. These factors, together with the introduction of an arbitrary grid size from which the vorticity is calculated, introduce an upper limit to the resolution of vorticity estimates.

Thus, all physical tracers may be seen to have severe limitations in application to periods exceeding a few days.

3.2. Gaseous tracers

Various trace gases are shown in table 3.2 and we briefly consider their suitability for use as tracers.

3.2.1. Water vapour

Water vapour is conservative in the absence of condensation and mixing, as perhaps in the stratosphere and, to a limited degree, in the upper troposphere.

The humidity sensors used in radiosondes are, without exception, inaccurate above the middle troposphere (Middleton and Spilhaus, 1955), because of their excessive lag at low temperature.

In the regions where water vapour is moderately conservative the temperatures vary within the range -40 to -90°C, and sampling may be carried out by the frost point hygrometer of Dobson et al (1946) and Mastenbrook and Dinger (1961); by infrared spectroscopy of the sun at various heights and differencing, either from a balloon borne platform (Murray et al, 1960), or from aircraft (Houghton 1962); and finally by direct sampling using cooled vapour traps (Barclay et al, 1960, Brown et al, 1961).

3.2.2. Ozone

Ozone is produced by photochemical processes by ultra-violet light as discussed by Chapman (1951). It has been shown fairly conclusively that ozone below 30 km is protected from photochemical dissociation by the ozone above that level (Nicolet, 1958, Dutsch, 1956, Craig, 1950). The 50% recovery times at 30 km are 3 days and a month for zenith and horizon sun respectively, and very much longer at lower levels. Thus, ozone is essentially conservative for periods of at least a few days near 30 km and for much longer below this height.

TABLE 3.2

GASEOUS TRACERS AND THEIR DEPARTURES FROM CONSERVATISM

Tracer	Source	Source Type	Sink	Sink Type
Water Vapour	Earth	Evaporation	1. Troposphere 2. Above 70-80 km.	Condensation Precipitation Photochemical decomposition
Ozone	Stratosphere	Photochemical reactions	1. Earth 2. Stratosphere above 30-35 km.	Reduction Photochemical decomposition
$C^{14}O_2$	Stratosphere	Nuclear explosions Cosmic ray 'stars'	1. Atmosphere 2. Ocean	Radioactive decay Solution

Ozone measurements fall into two categories:

1. Total ozone in a vertical column above the station, (this is a crude form of tracer since most of it occurs in the stratosphere below the 30 km level).

2. Vertical profiles of ozone concentration or mixing ratio. Measurements of total ozone are made using the Dobson spectrophotometer on the sun, zenith sky either clear or clouded, and the moon as radiation sources. The method utilises the differential absorption by ozone in two neighbouring wavelengths $\sim$ 3,200 Å as described comprehensively by Dobson (1957).

Measurement of profiles may be made either by remote sampling by surface based instruments, or by direct sampling instruments carried aloft by balloons, aircraft or rockets.

The most used remote sampling method is the 'Umkehr' method which makes use of the differential absorption of ultra-violet from zenith sky light by ozone at various solar zenith angles. The technique has been described and methods recommended for interpreting the observations by Ramanathan and Dave (1957), Dütsch (1959), Mateer (1960).

The profile may also be estimated for layers of finite depth from emission measurements in the 9.6 μ infra red band. The method was described by Goody and Roach (1958), and has since been modified and critically appraised by Walshaw (1960), and Ooyoma (1962).

Direct observations fall in two subgroups - those measuring the total ozone above the instrument at any instant in its flight and differencing, and those directly sampling the ozone in the air in the environment of the sampling device.

The former method, initiated by E. and V. Regener (1934) makes use of the differential absorption of ultraviolet by ozone. It has been used from Balloons (Regener, 1951), and rockets (Johnson et al, 1954), Hulbert, 1955), and has been refined for use on a radiosonde by Paetzold (1954).

Environmental sampling may make use of the oxidation of potassium iodide by ozone (Regener, 1959), Brewer and Milford, 1960), or the oxidation luminescence device of Regener (1960).

Comparison tests of different instruments (Moreland, 1959, and Brewer et al, 1960) show considerable differences in simultaneous profiles from different types of instrument but good qualitative agreement between instruments of the same type.

3.2.3. Carbon-14 isotope in carbon dioxide

C^{14} is produced from N by cosmic rays and by nuclear explosions, and becomes attached to form $C^{14}0_2$. Its rate of production by cosmic rays is closely proportional to the number of cosmic ray stars which is a function of latitude and height (fig. 3.1), but independent of time. Since the half life of C^{14} is 5,600 years it is conservative with respect to decay.

A method of direct sampling has been used whereby a large volume of air is collected at a standard level compressed and analysed in the laboratory (Hagemann et al, 1959).

3.3. Particulate tracers

Among possible particulate tracers table 3.3 shows a selection of those with most obvious application.

3.3.1. Aerosol particles of radius $< 0{\cdot}1\,\mu$

Both the effects of sedimentation and coagulation place limitations on the use of particulates as tracers.

Figure 3.2 from Junge et al (1961) shows the gravitational settling rates for spherical particles, calculated from the Stokes-Cunningham formula. We observe that the sedimentation may be neglected for particles whose radius is below $0.1\,\mu$ since the settling rates are less than vertical velocities commonly observed in the lower stratosphere. Junge et al (1961) also estimate the half life of particles as a result of coagulation with a background population of larger particles - self coagulation being neglected.

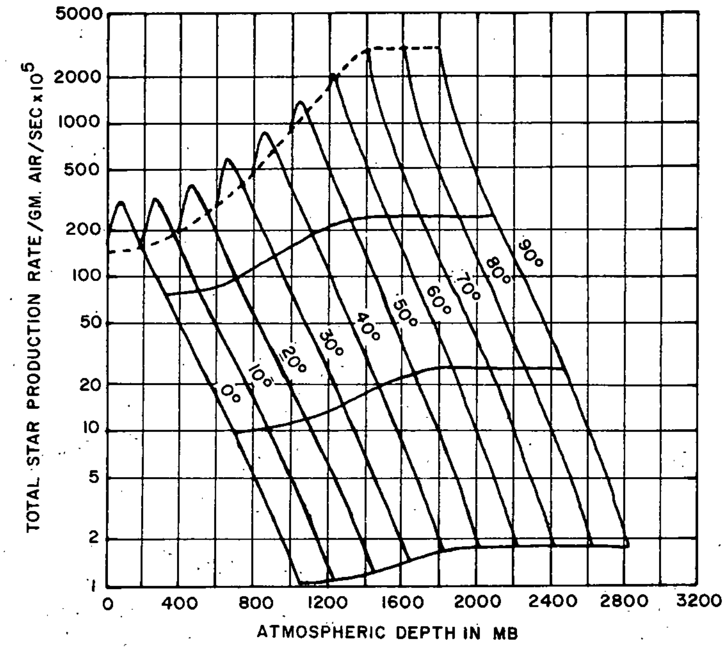

Figure 3.1.
Star Production Rate as a Function of Pressure, Curves for Successive 10° lat. Intervals are Displaced 200 Units Along the Abscissa

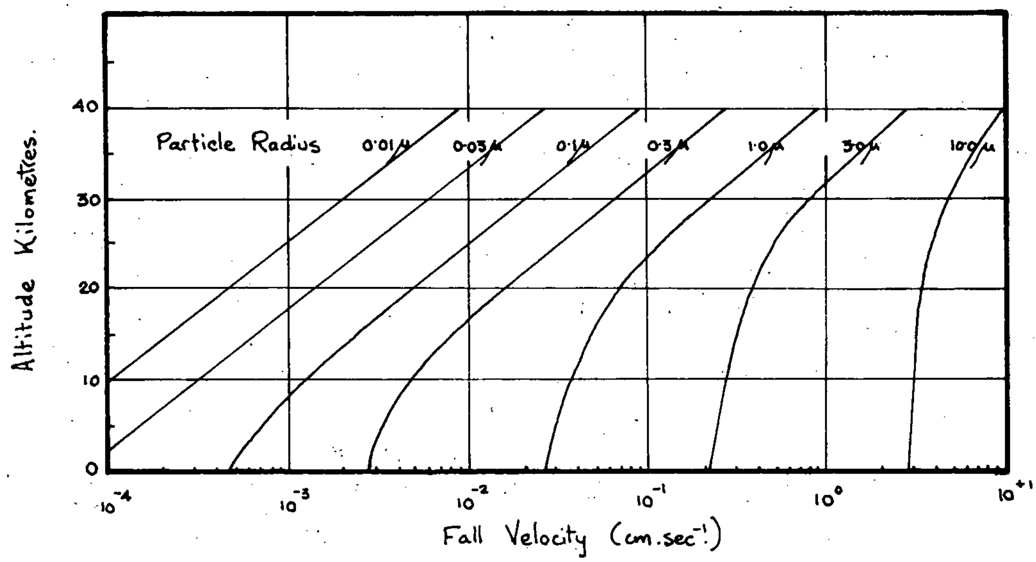

Figure 3.2.
Velocity of Gravitational Settling for Spherical Particles of Density 2 gm. cm^{-3}.

TABLE 3.3

PARTICULATE TRACERS AND THEIR DEPARTURES FROM CONSERVATISM

(Half Lives of Radioisotopes are Shown in Brackets)

Tracer	Source	Source Type	Sink	Sink Type
Aerosol Particles of radius $< 0.1\ \mu$	Troposphere	Uncertain	1. Stratosphere 2. Earth	Coagulation Wet, Dry deposition
Bomb produced radioisotopes S_r^{89} (so 4d), Z_r^{95} (65d), W^{185} (74d), R_h^{102} (210d), S_r^{90} (28y), C_s^{137} (30y)	Atmosphere	Nuclear explosions	Earth	Wet, Dry deposition

The results in table 3.4 show that coagulation may be neglected in the size range 0.01-0.1 μ .

Vertical profiles of sub-decimicron particles are obtained using a balloon borne automatic recording Aitken nuclei counter with a chamber pressurisation device described by Junge et al (1961).

3.3.2. Artificially produced radioactive isotopes

In addition to considerations of gravitational sedimentation and coagulation, radioactive isotopes depart from conservatism as a result of radioactive decay. Their sporadic injection in time and locality further complicates the interpretation of their four dimensional distribution.

In each nuclear explosion radioactive debris, produced by vaporization and irradiation of the environment results, on subsequent recondensation, in an initial particle concentration spectrum probably heavily weighted toward small radius. In megaton bursts these particles are injected into the stratosphere where quasihorizontal winds distribute them approximately in a zonal belt around the hemisphere and gravitational settling out of larger particles takes place.

As a result of the increase of sedimentation velocity with height (figure 3.2) at any instant the spectrum of debris concentration will be more biased towards larger radius with increase in pressure. This may result in a departure from conservatism, unless we limit sampling throughout the stratosphere to the same particle size range, and may aid in interpreting results of radiodebris sampling.

If the half life is long compared with the period of the motion being studied, radioactive decay may be neglected, if not, provided the isotope concentration has not recently been disturbed by a fresh injection, the decay may be removed for a specific isotope by normalizing all activities to a fixed time.

Particulates are removed from the atmosphere both by wet and dry deposition but the physical processes are not clearly understood (Greenfield, 1957, Small, 1960, Itagaki and Koenuma,

TABLE 3.4

HALF LIVES OF PARTICLES OF VARIOUS RADII IN DAYS AS A RESULT OF COAGULATION WITH THE BACKGROUND POPULATION OF PARTICLES FOR VARIOUS ALTITUDES

Altitude in km. \ Radius in μ	0.0025	0.005	0.01	0.02	0.04
27.5	1.6	6.4	24.	90.	340.
20.0	5.1	20.	80.	280.	1060.
12.5	17.	61.	210.	740.	2570.

1962), and the relation of strength of sink to concentration is correspondingly lacking in precision.

Radioactive isotope concentrations are, in general, so minute that sampling requires time intervals of hours to weeks.

Deposition measurements have been made either by collection of the total deposition over a given area, including the precipitation during the period, or by exposing a one foot square gummed film 3 feet above the ground for 24 hour periods. The rainfall during the period washing over the film not being collected. Welford and Collins (1960), by analysing simultaneous samples from the various types of collectors, viz. tub, pot and funnel, together with gummed film, concluded that the various collection type samplers gave consistent results but there was no obvious correlation between them and the collection by gummed film.

Air concentrations are measured by passing a considerable volume of air through a filter at the earth's surface, on balloons (U. S.) and on aircraft flights (U. S. and U. K.). Attempts were also made to measure surface air activity by means of one foot square cheesecloth screens mounted in a vertical plane normal to the airstream, but Lockart et al (1959) found little correlation with simultaneous sampling by the standard filter technique.

We suggested earlier that conservatism may only be a plausible assumption for particulate tracers if a limited size range is collected at all levels. In fact, as we observe from Holland's (1959) effeciency height plots (figure 3.3) the millipore filters commonly used remove the largest particles with highest efficiency, and the millimicron particles, which form the bulk of the population, with the least. We presented plausible reasons that the activity may be concentrated in larger mean particle size with increasing pressure and the variation in collection efficiency may further accentuate this nonconservative effect.

4. A Climatological Analysis of Tracer Distributions

The various tracers will be analysed, where possible, in terms of meridional and zonal distributions, either for the year

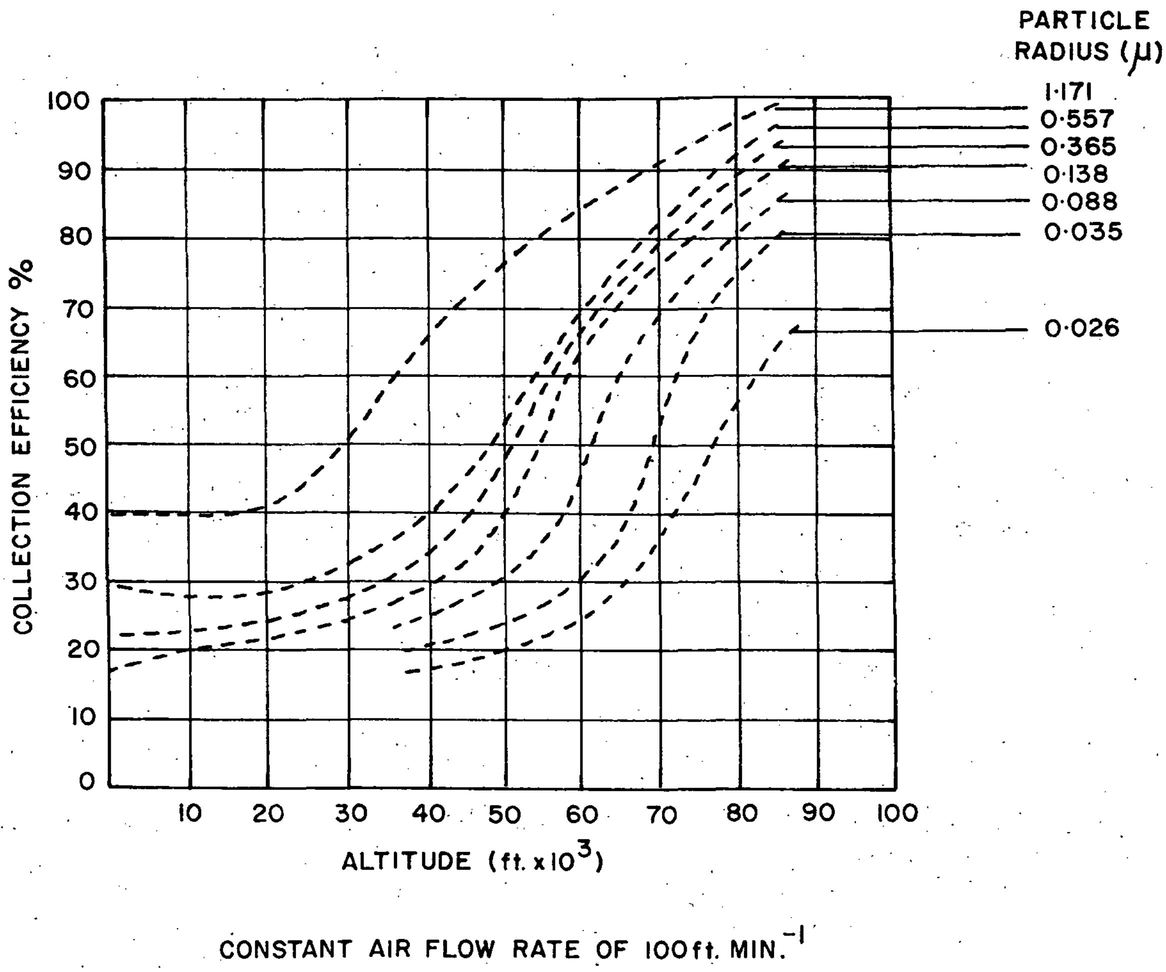

Figure 3.3.
Variation of Collection Efficiency with Particle Size
for Porous Filter of General Mills Inc. Design. (After Holland 1959).

or season. Some correlations of total ozone with features of the temperature field will be examined in investigating eddy transfer in the lower stratosphere.

4.1. Water vapour

A summary of the water vapour observations in the upper troposphere and stratosphere is shown in tables 4.1 and 4.2. From flights listed in table 4.1 between the equator, England and Iceland meridional sections for winter and summer were constructed in figure 4.1.

The observational sample is limited but should give a reliable qualitative description of the distribution in low latitudes. In high latitudes there was a period in January and February, 1962, when much higher frost points than usual were observed during Met. Research Flights from Leuchars, Scotland. The approximately fivefold increase in mixing ratio suggests the observations might have been unreliable, but they are not inconsistent with other mixing ratios in the lower stratosphere at lower latitudes. Moreover, Tucker, (1957), evaluated mean profiles of frost point for observations in various tropopause height ranges over Southern England and showed frost point at a given level in the lower stratosphere to range over 15°K with changes in tropopause height, and it is to be expected that this variation will be at least as noticeable in higher latitudes.

No information is currently available on zonal variations in water vapour.

The series of observations up to the middle stratosphere in table 4.2 are restricted to a 15° latitude belt about 45°N and between 0° and 105°W. In general, the humidity mixing ratio appears to fall with height to a level between 150 and 200 mb, then increase once more to a value, at a height of 50-25 mb, about an order of magnitude greater than the mean value at the tropopause over England. Mastenbrook (1963), has shown, by comparing ascent and descent vapour profiles in August, September, October and April over the U. S., that the apparent increase in mixing ratio in middle stratosphere probably results from contamination from the balloon, and casts reasonable doubt on all balloon observations. He shows

TABLE 4.1

OBSERVATIONS MADE WITHIN THE TROPOSPHERE AND LOWER STRATOSPHERE BY INSTRUMENTS MOUNTED IN AIRCRAFT OF THE BRITISH METEOROLOGICAL RESEARCH FLIGHT

Observation Reference		Locality	No. of Flights	Season	Observational Method
Bannon et al.	(1952)	Vicinity of England	130	all	
Murgatroyd et al.	(1955)	Vicinity of England	35	all	Frost point hygrometer
Goldsmith	(1954)	Sudan		Summer	mounted in Meteorological
Helliwell et al	(1956)	Vicinity of England	46	all	Research Flight aircraft.
Tucker	(1957)	Vicinity of England	399	all	
Helliwell & Mackenzie	(1957)	Farnborough south to Idris	17	Summer	
Helliwell	(1960)	40° to 67° N.		Spring, Summer	
Kerley	(1961)	Malta to Niarobi	12	Summer	
Houghton & Seeley	(1960)	England and Malta	7	Winter	Solar spectrometer in aircraft.

TABLE 4.2

OBSERVATIONS OF VERTICAL PROFILES, AND VALUES AT ABOUT 30km. MADE WITH BALLOON-BORNE INSTRUMENTS

Observation Reference	Latitude	Longitude	No. of Profiles	Season	Observational Method
Barrett et al (1950)	37°N	100° W	3	1.7-49, 26.8-49 7.1-50.	Dewpoint radiosonde.
Brasefield (1954)	40°N	75° W	> 7	All	Dewpoint radiosonde.
Mastenbrook & Dinger (1960)	U.S.A.		11	All	Dewpoint radiosonde.
Mastenbrook & Dinger (1961)	40°N 39°N	105° W 75° W	1 2	28.4-59. 8.4-60, 27.6-60	Dewpoint radiosonde.
Murcray et al (1960)	40° N	105° W	1	19.6-59.	Solar spectrometer.
Murcray et al (1962)	40° N	105° W	1	18.5-60.	Solar spectrometer.
Barclay et al (1960)	52° N	0° W	1	2.5-58.	Vapour trap.
Brown et al (1961)	52° N	0° W	9	Summer Autumn.	Vapour trap.

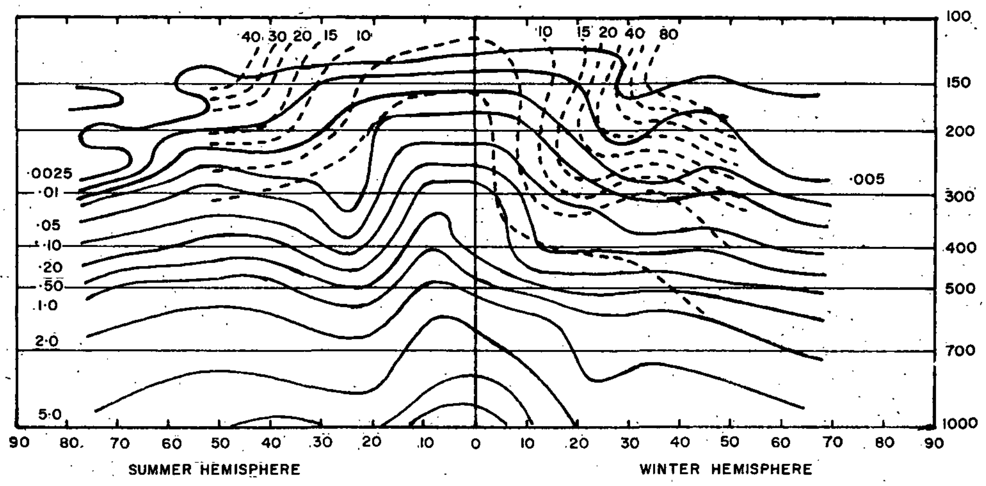

Figure 4.1.
Mean Meridional Sections of Ozone and Humidity Mixing Ratio

-----	Ozone Volume Mixing Ratio	(Units of 10^{-8})
———	Humidity Mixing Ratio	(Units gm kgm^{-1})

the mixing ratio to vary between .01 and .04 gm kg^{-1}, to at least 30 km. The results of Houghton and Seeley (1960), using a spectroscopic method appear consistent with the concept of a dry middle stratosphere.

4.3. Ozone

In the following treatment we will be concerned with total ozone, 0_{Σ}, in the vertical, and with vertical profiles of mixing ratio, X_{op}, at pressure p.

4.3.1. Total ozone

The meridional and zonal variations in the total ozone amount, 0_{Σ}, are best illustrated by London's (1962) seasonal northern hemispheric analyses in figures 4.2 to 4.5. The ozone amount is observed, at almost all longitudes and seasons, to increase with latitude to 50-80°N, then remain constant or decrease slightly northwards to the pole. In section 5.1 the zonal variation in ozone will be shown to be related to the position of the standing eddies.

4.3.2. Vertical profiles of ozone and three dimensional distributions

Figures 4.1 and 4.2 show tentative seasonal meridional sections of ozone constructed from data collected by the British Met. Research Flight using a modified Brewer type chemical sampler.

A summary of ozone vertical profile measurements is presented in table 4.3 and an extensive series of Regener sonde profiles over the U. S. have been accumulating since the I. G. Y. The inconsistency between ozone profiles measured by different devices was illustrated by the intercomparison tests of Brewer et al (1960), and is even observed in 'Umkehr' profiles derived using different computation techniques, (Dutsch, 1959). Figure 4.6 shows representative summer and winter cross sections in which the cross sections of Ramanathan and Kulkarni (1960) have been modified to incorporate the mean profiles for Churchill and Moosonee from Mateer and Godson (1960) thus, introducing the winter secondary maximum which is not nearly so prominent over European stations. The concentrations have been converted to mixing ratios below 30 km in figure 4.7.

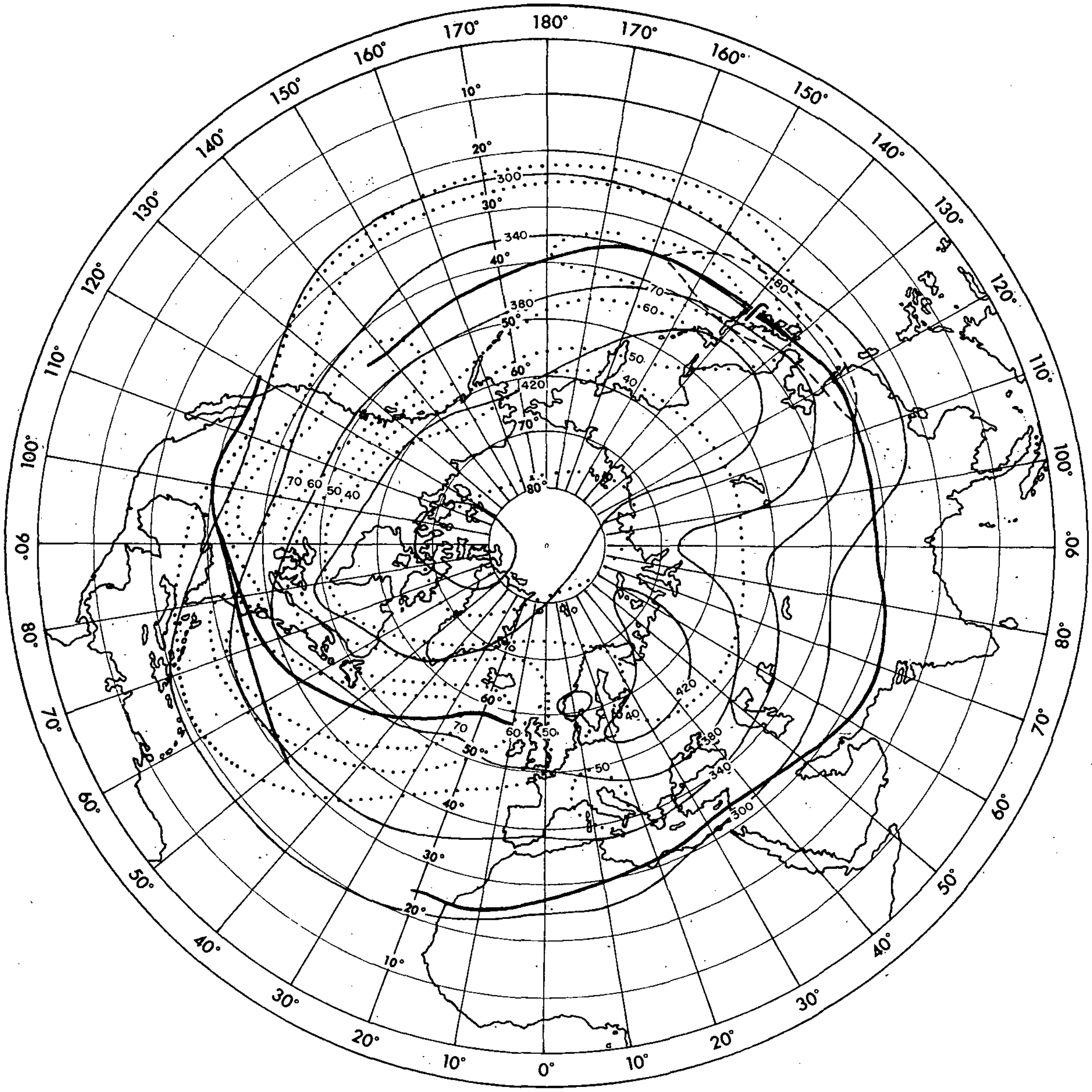

Figure 4. 2.
The Distribution of 3 Monthly Mean Values of Total Ozone in Spring, and the Annual Sr^{90} Activity Concentration are Shown, Together with the Axis of Maximum Wind from the Calculated Seasonal Mean Values of Crutcher (1962)

———	Total Ozone (cm. 10^3 S. T. P.)
·····	Sr^{90} Activity Concentration
━━━	Axis of Maximum Wind
- - - - -	Isotachs

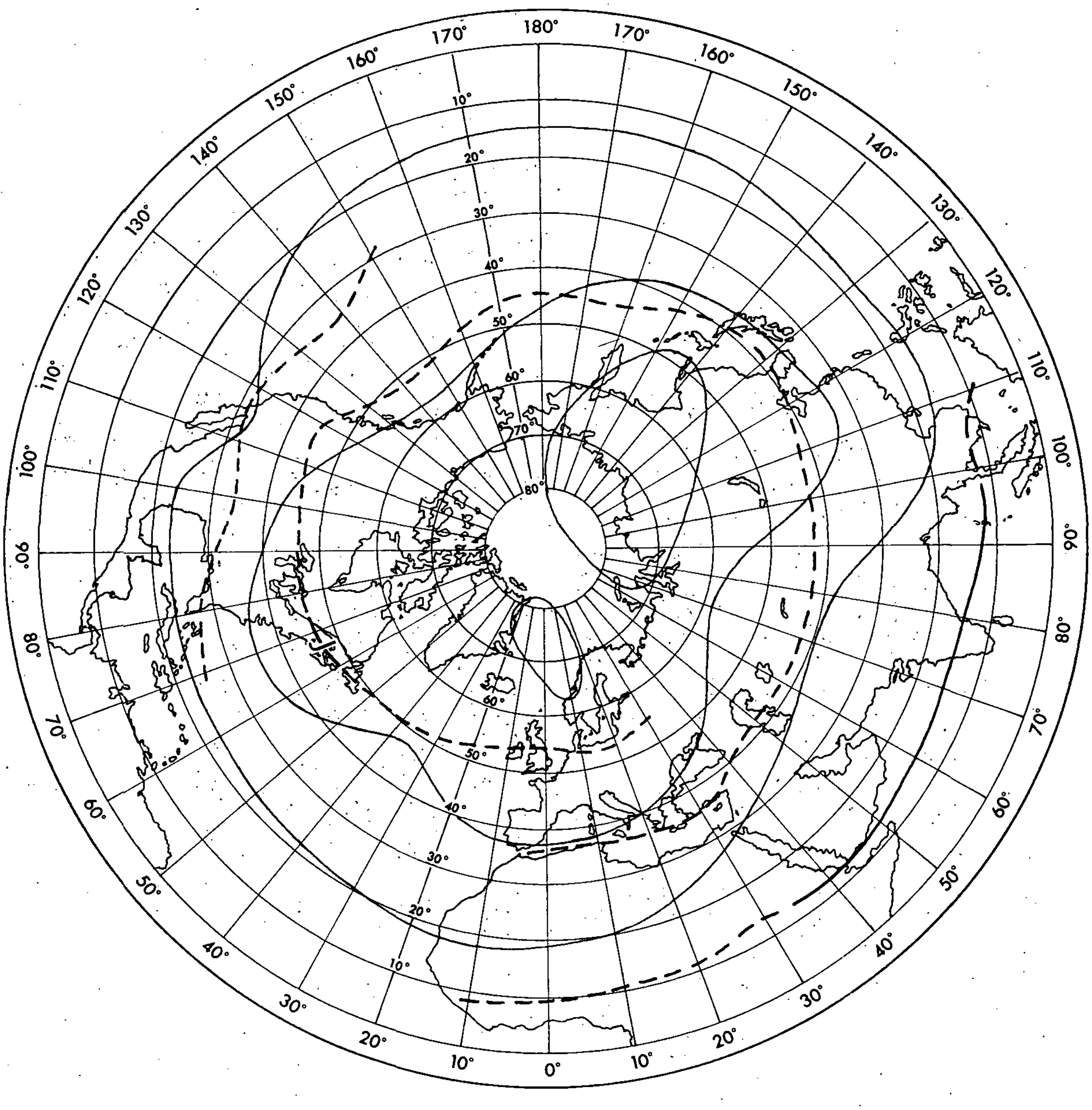

Figure 4.3.

The Distribution of 3 Monthly Mean Total Ozone for Summer, (London 1962), is Shown Together with the Axis of Maximum Wind from the Seasonal Mean Values of Crutcher 1962

——— Total Ozone (cm 10^3 S.T.P.)

━━━ Axis of Maximum Wind

- - - - Line Indicates Mean Wind is Less Than 60 Knots

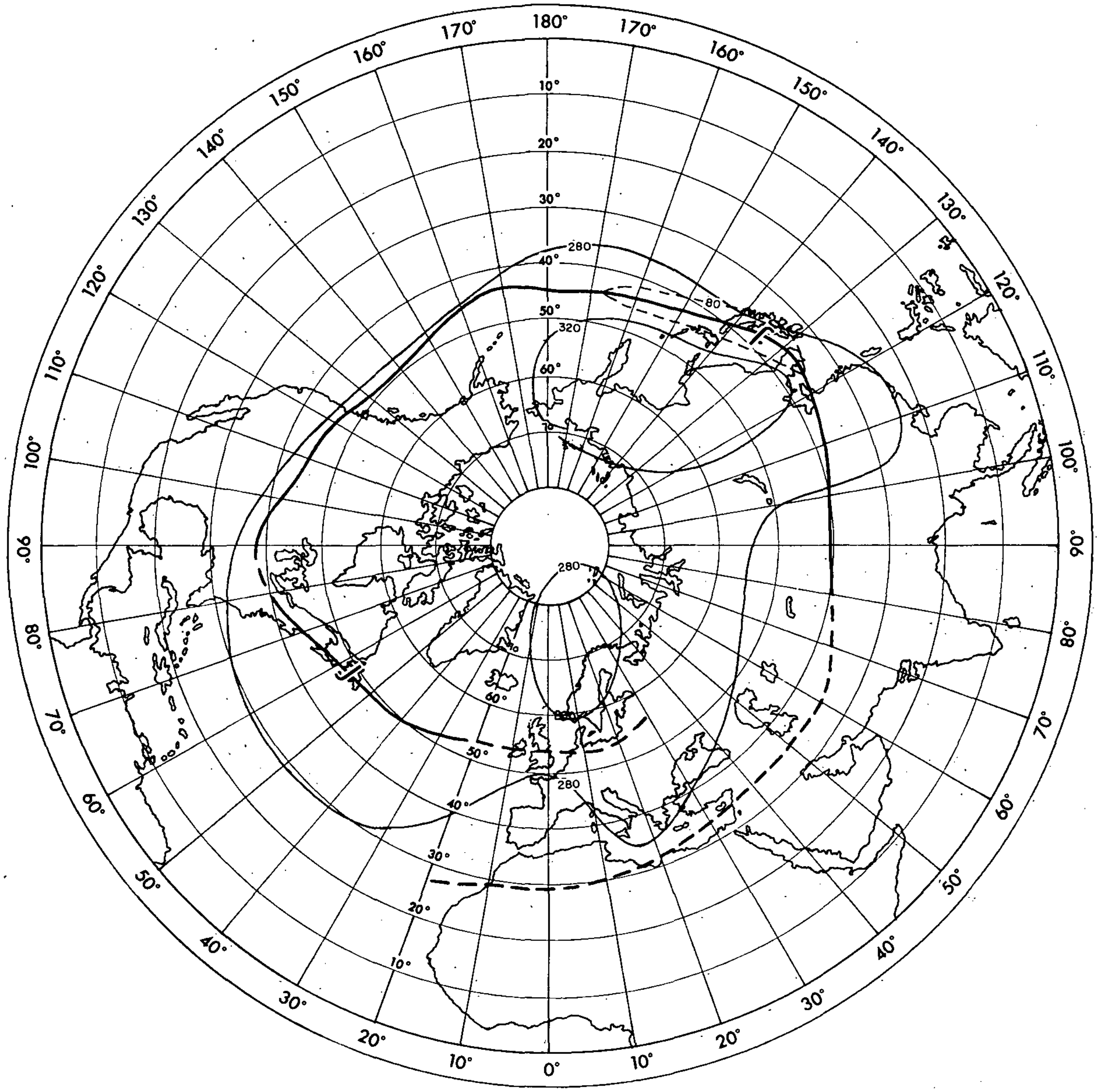

Figure 4.4.
The Distribution of 3 Monthly Mean Values of Total Ozone in Autumn, (London 1962), is shown Together with the Axis of Maximum Wind From the Seasonal Mean Values of Crutcher 1962.

———	Total Ozone (cm 10^3 S.T.P.)
▬▬▬	Axis of Maximum Wind
- - - - -	Isotachs

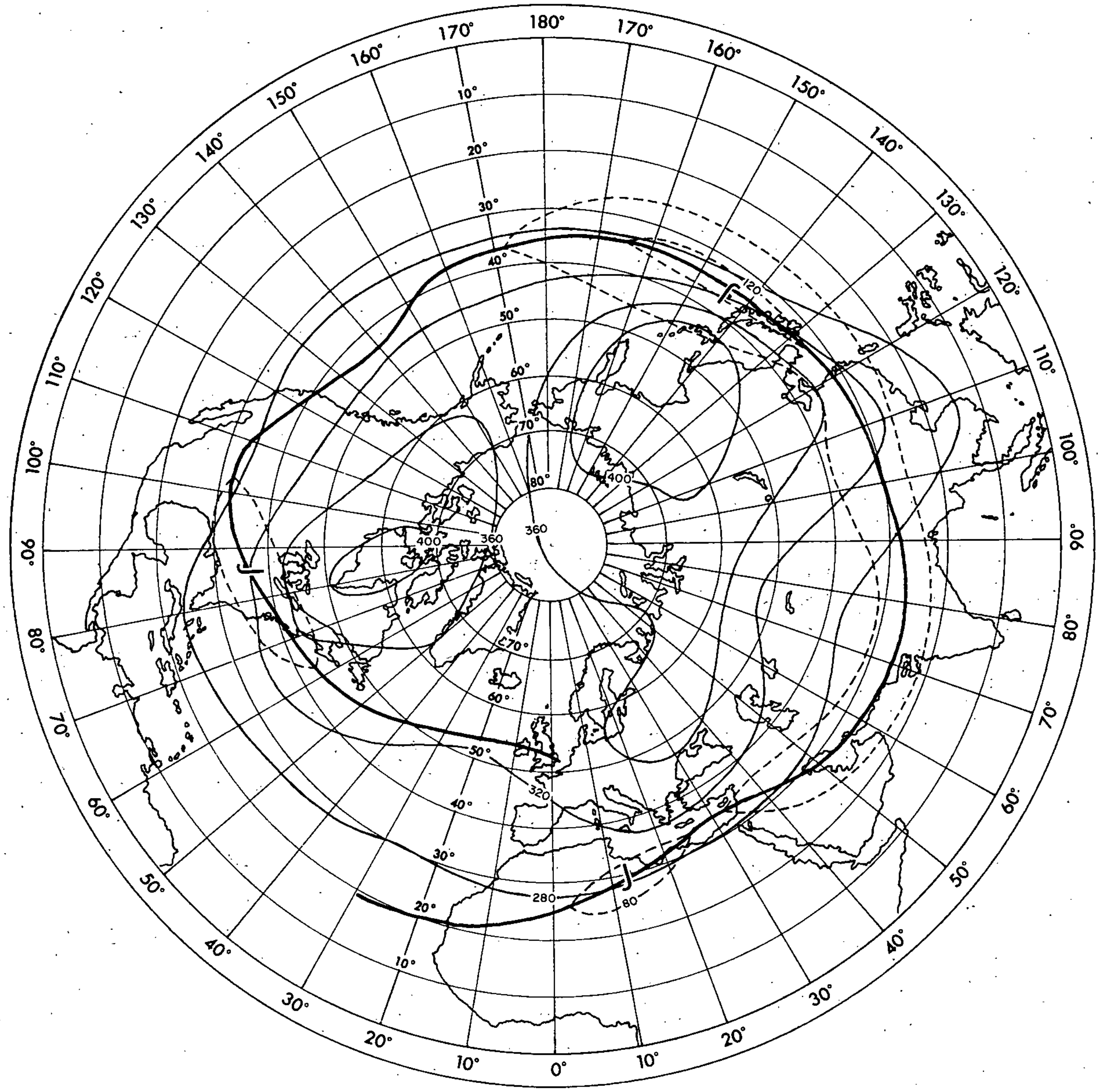

Figure 4. 5.
The Distribution of 3 Monthly Mean Values of Total Ozone in Winter, (London 1962), is shown Together with the Axis of Maximum Wind From the Seasonal Mean Values of Crutcher 1962.

———	Total Ozone (cm 10^{-3} S. T. P.)
▬▬▬	Axis of Maximum Wind
- - - - -	Isotachs

TABLE 4.3

OBSERVATIONS OF VERTICAL PROFILES OF OZONE
SPECIFYING THE MEASUREMENT TECHNIQUE USED IN EACH CASE

Observation Reference		Approximate Location	No. of Observations	Season	Observational Method
Gotz, Meetham & Dobson	(1934)	47°N, 10°E	46	All	'Umkehr' method.
E. & V. Regener	(1934)	37°N, 100°W	1	Summer	Spectrograph ascent in an unmanned balloon.
Meetham & Dobson	(1935)	70°N, 19°E	13	Summer	'Umkehr' method.
O'Brien et al	(1936)	44°N, 100°W	1	Winter	Spectrograph in manned balloon - Explorer II.
Tonsberg & Olsen	(1944)	70°N, 19°E	64	All but Autumn	'Umkehr' method.
Coblentz & Stair	(1939)	39°N, 77°W	4	Summer	Ultra-violet intensity meter in unmanned balloon.
Coblentz & Stair	(1941)	39°N, 77°W	19	All	Ultra-violet intensity meter in unmanned balloon.
Newell & Siry	(1947)	37°N, 105°W	1	Winter	Spectrograph in rocket.
Ramanathan & Karandikar	(1949)	28.5°N, 77°E 18.5°N, 73.9°E		All	'Umkehr' method.
Karandikar	(1952)	28.5'N, 77°E 18.5'N, 73.9°E		All	'Umkehr' method.
Ramanathan	(1953)	10-30°N, 75°E		All	'Umkehr' method.
Hulbert	(1955)	37°N, 105°W	7	All	Spectrograph in rockets.
Paetzold	(1953)	Collected Profiles for European Stations (Bibliography)			'Umkehr' method.
Paetzold	(1955)	51°N, 10°E	17	All	Ultra-violet intensity sonde.
Paetzold, Piscalar	(1961)	4°S, 48°N; 70°N, 0°W	8	Spring, Winter	Optical sonde.
Dutsch	(1959)	47°N, 10°E	30	All	'Umkehr' method.
Brewer & Milford	(1960)	55°N, 5°W	32	All	Chemical sonde.
		70°N, 19°E	3	Autumn	Chemical sonde.
		47°N, 10°E	12	Autumn	Chemical sonde.
Godson & Matteer	(1960)	51°N, 81°W 74°N, 95°W	27 4	All	'Umkehr' method.
Kulkarni	(1962)	38°S, 145°E	15	All	'Umkehr' method.

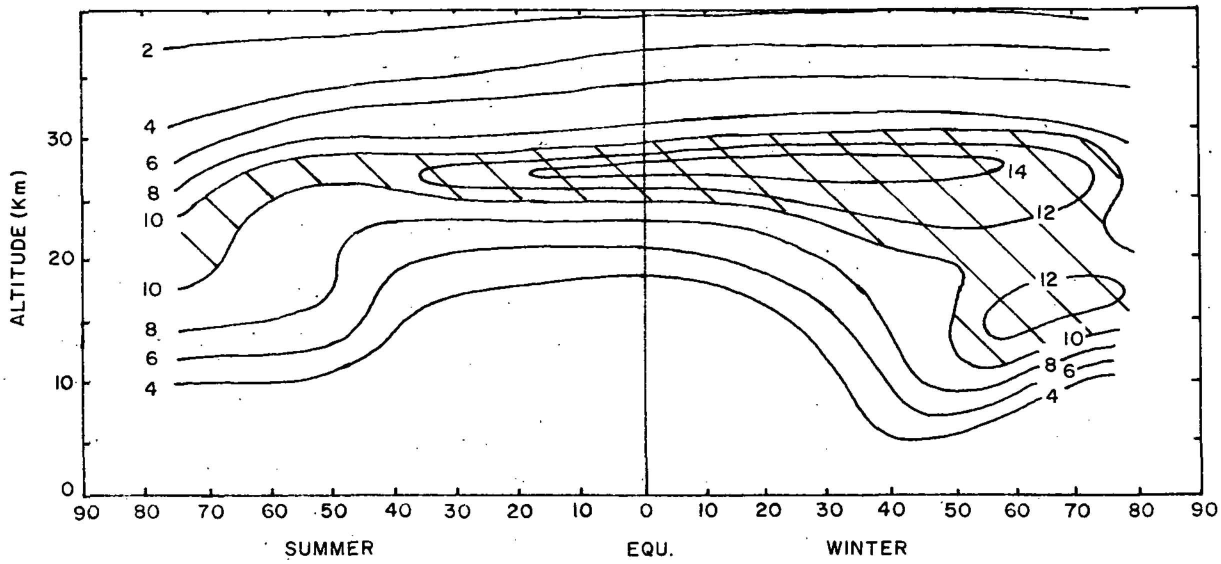

Figure 4. 6.
Representative Meridional Sections of Ozone Concentrations
(10^{-3} cm·km^{-1})

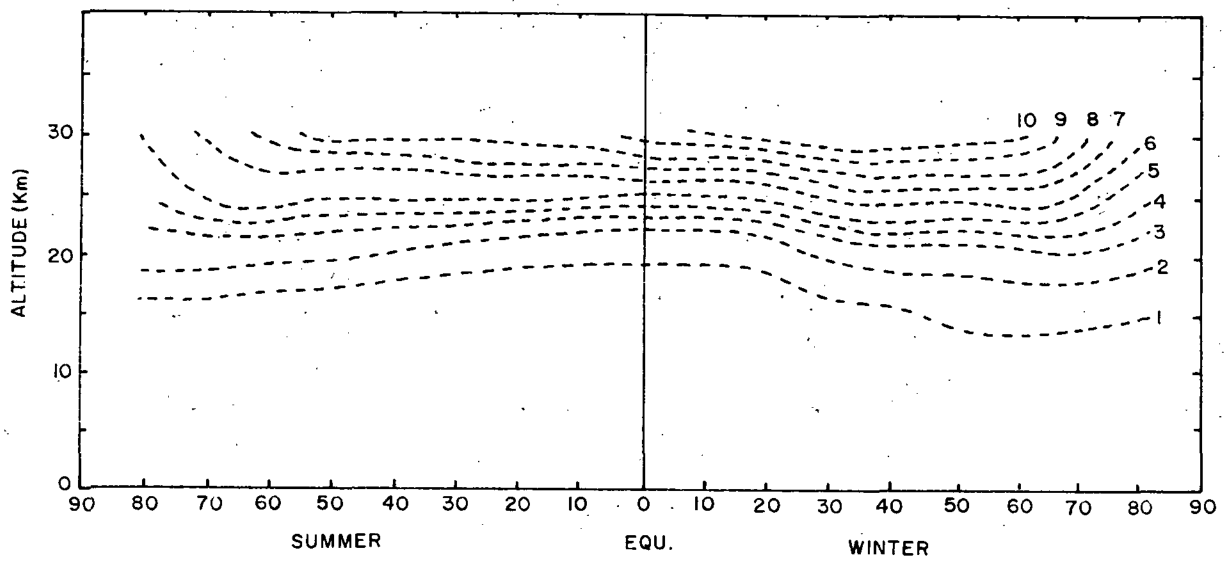

Figure 4. 7.
Meridional Cross Sections of Ozone Mixing Ratio Derived from the above Concentrations and Temperatures from Murgatroyd (1957).

4.4. Radioactive isotopes from Nuclear weapons

The irregular distribution of nuclear explosions in space and time makes special caution necessary in seeking the meteorological significance of observed distributions of the isotopes produced. It will, however, appear that certain distributions are unquestionably of high meteorological significance.

4.4.1. Activity at and near the ground

Both air and rain samples may be evaluated for total activity or better for individual isotopes. Unfortunately, the lack of consistency in sampling by the various agencies creates difficulty in synthesizing the results into a useful four dimensional distribution.

The meridional distribution of radioactive fallout over a year may be estimated either by soil sampling or, if dry deposition is negligible, from rain sampling. The former method, used extensively since 1955, as reported by Martell (1959) and Alaxander (1960), while not a particularly good technique has the advantage of being the one method from which rapid estimates of the world-wide deposition may be made, without an extensive programme of instrument standardisation. For this reason no attempt has been made to tabulate the extensive data sources but only to present consistent data analysis.

We observe a similar pattern (figure 4.8) of meridional deposition of S_r^{90} obtained from soil sampling in successive years (Alexander, 1960), and from measurements of S_r^{90} in rain (Stewart et al, 1959), - a minimum at the equator bounded by distinct maxima in the 30-50^c latitude belt in both hemispheres with values decreasing thereafter with latitude to at least 70°N and 60°S.

The meridional variation of mean annual rainfall is shown as a histogram in figure 4.8. The mid-latitude maximum corresponds closely with that of deposition but the high tropical rainfall has no obvious counterpart in the deposition pattern. Thus, the deposition is not solely a function of the rainfall amount, and if we are to

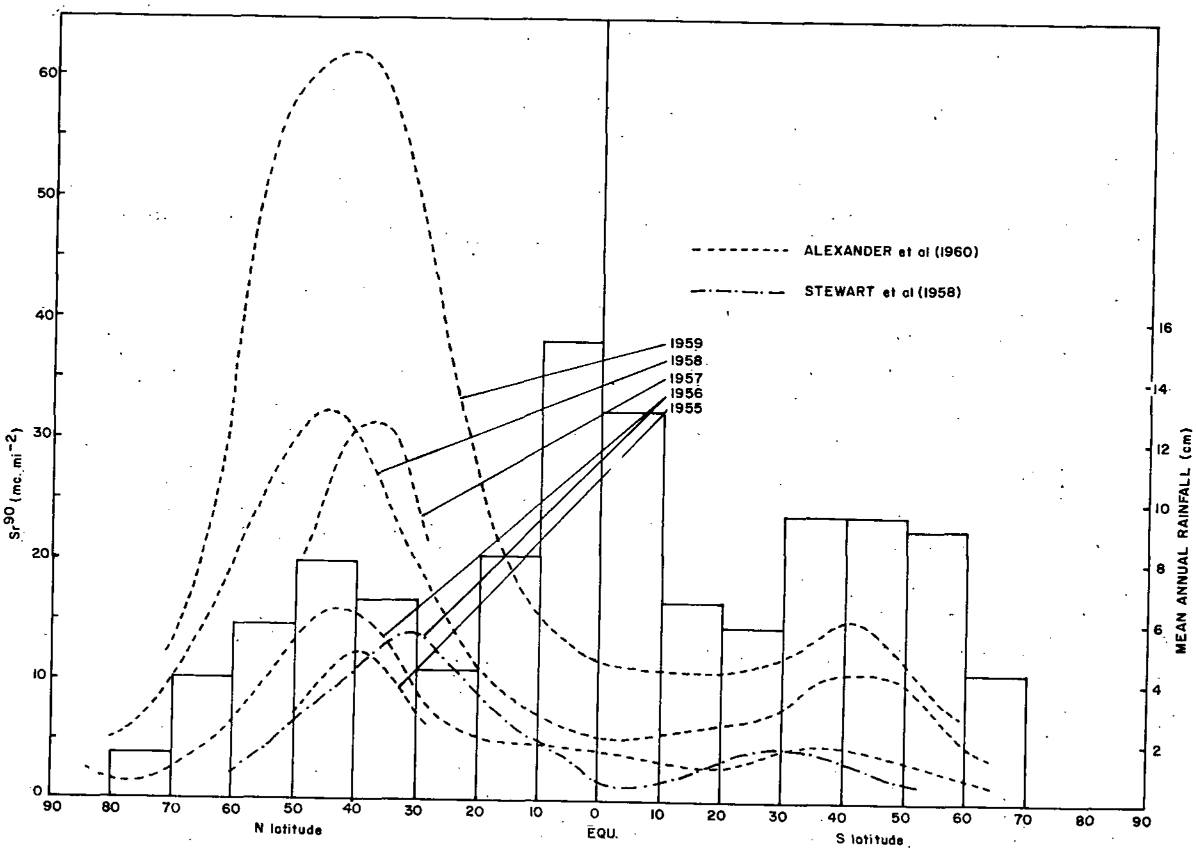

Figure 4.8.
The Continuous Curves show Sr^{90} Deposition in Successive 1 Year Periods from 1955-1959 and the Histogram shows the Meridional Variation of Mean Annual Rainfall

interpret these and other deposition results unambigously in terms of activity in air, we must detect the relation between them.

The meridional variation of annual values of S_r^{90} concentration in rainwater in figure 4.9 for different sets of years from Stewart et al (1959), and Crooks et al (1960), shows a minimum in the tropics, a maximum near 40° N, a decrease to 60-70°N and a final increase towards the pole.

The empirical studies of Stewart et al (1959), Crooks at (1960), Bliechrodt et al (1959), Storeb (1959), Small (1960) and Alexander (1960), are in general agreement with Stewart (1958), that the monthly mean values of specific activity in rainfall are approximately proportional to the concentration in the air through which it falls, so that the former can be taken as a climatological measure of the latter, except in low latitudes. The close correspondence in the meridional and seasonal variations in the gross fission product activity in surface air along 80° W (Lockhart et al, 1959, 1960 a, b), and in specific activity in rain (Staley, 1962), in figure 4.10, provide further confirmation of this relation.

We observe the position of the meridional mid-latitude maximum of both properties exhibits a seasonal N-S fluctuation.

The concentration S_r^{90} in rain at mid-latitude stations from 1954-1962 is shown in figure 4.11 (from Crooks et al, 1961), 1962). A seasonal variation with strong spring maximum and autumn minimum is evident throughout the period and is apparently little influenced by the incidence of injections. From the inventory of nuclear explosions presented graphically at the top of figure 4.10, we see that there were no high yield stratospheric bursts in the years 1955, and 1959-1961, yet the spring maximum remained obvious in each year.

4.4.2. Activities in the free atmosphere

Two specific radioisotopes were injected during the testing schedule as detailed below:

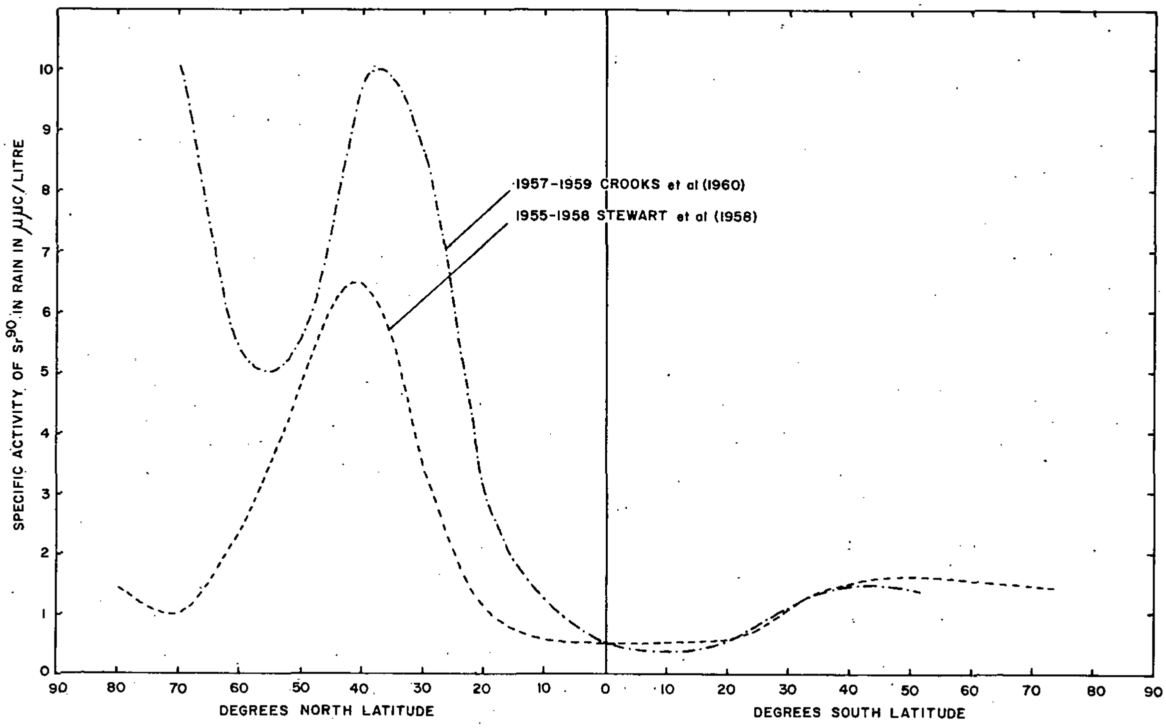

Figure 4. 9.

Meridional Variation of Sr^{90} Concentration in Rainwater

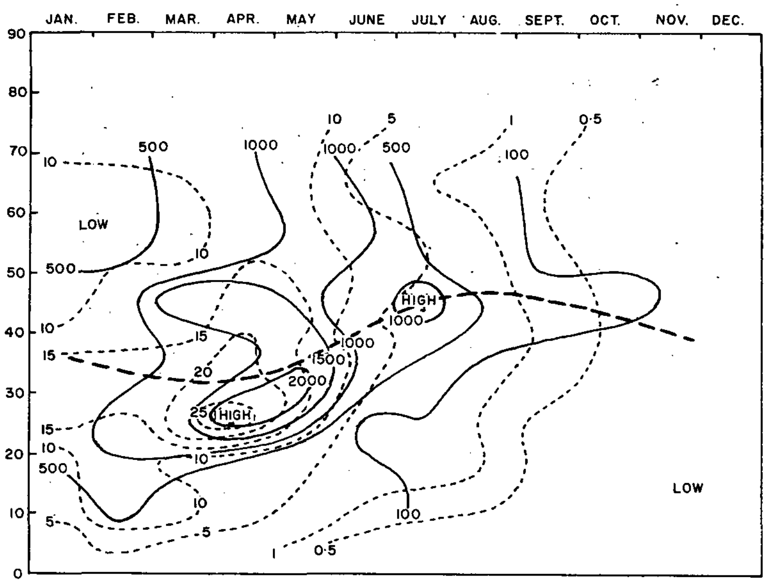

Figure 4. 10.

----- Time-Latitude Section of Gross Fission Product Activity in Air From Data Published by Lochart et al (1959, 1960)

Units: Disintegration per Minute per Cubic Meter.

_____ Time-Latitude Section of Rain Fission Product Content from Staley, (1962) (units μ C. M^{-2} per Inch of Rain)

— — — Position of Mean Zonal Wind Maximum at 80° W from Crutcher, (1961)

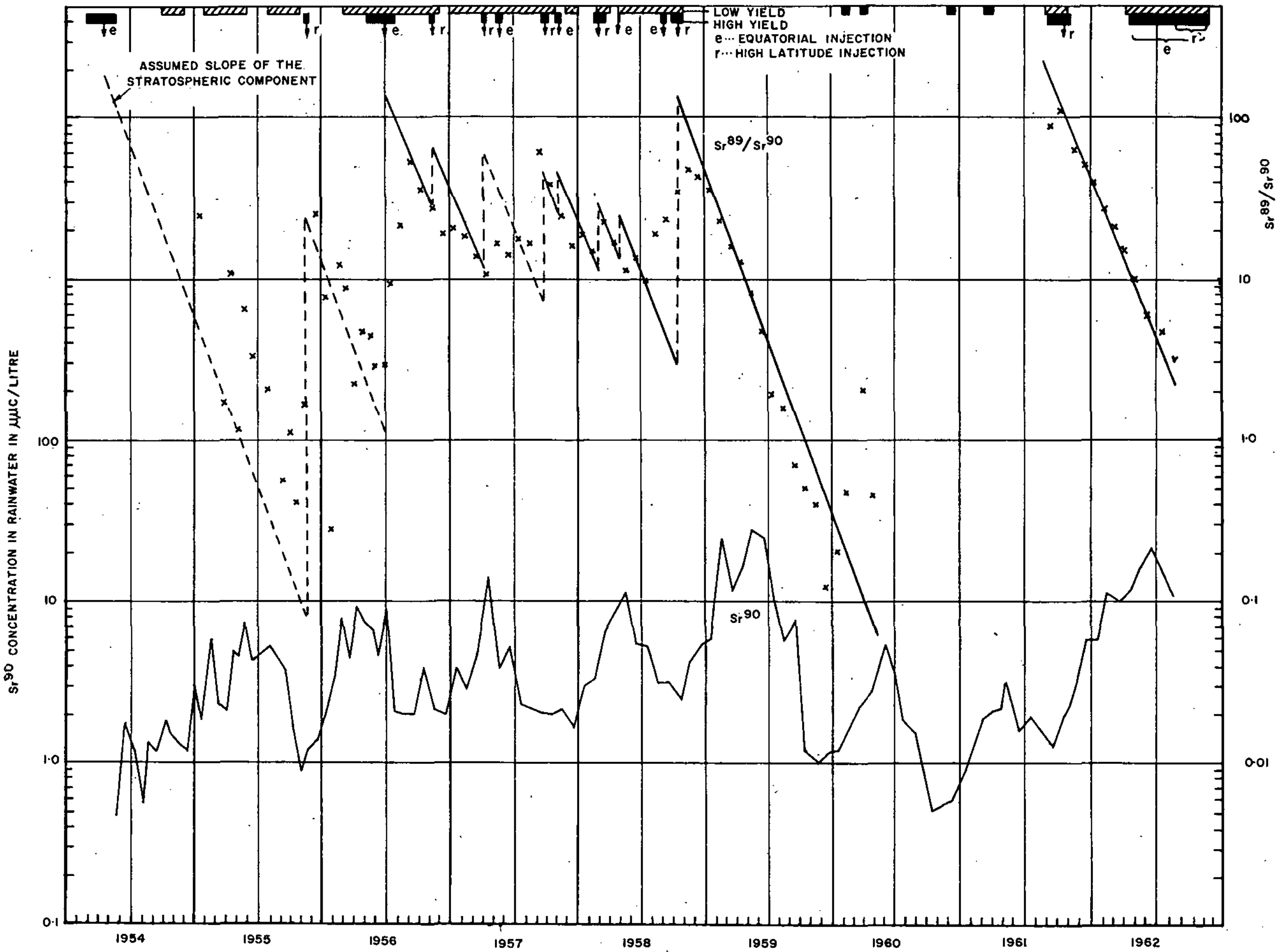

Figure 4. 11.
Time Series of Monthly Mean Values of Sr^{90} Concentration and Sr^{89}/Sr^{90} Ratio in Rainwater at Milford Haven (South Wales).
Schedule of Injections is Shown at the Top.

1. Tungsten 185 isotope, W^{185}, was injected at a height close to 25 km. during the Hardtack series of tests at 12°N in summer 1958.

2. Rhodium 102, Rh^{102}, was injected above 30 km during the Orange test at 17°N latitude in August, 1958.

They were sampled later in the stratosphere from aircraft and balloons, together with S_r^{90} which was injected in all tests, and their distributions are shown in figures 4.12-4.17. Distributions of excess C^{14} from balloon sampling of CO_2 by Hageman et al (1959), are shown in figures 4.18-4.19. We observe the following features:

1. Where a maximum exists in the vertical profile the height of this maximum varies little with time, and rises toward the equator.

2. Concentrations are barely detactable in the tropics to a height of 16 km.

3. The tropical lower stratosphere appears to be relatively poor in W^{185} and excess C^{14} in winter compared with summer.

5. Tracer Distributions with Respect to Synoptic Features

The obvious lack of uniformity in tracer distributions observed in the preceding section may be used to elucidate atmospheric transfers either climatologically by noting associations between the tracers and corresponding features of the fields of winds and temperature; and establishing correlations between such parameters, or by individual case studies.

5.1. Association in the mean distributions with respect to axes fixed on the earth's surface

In the cross section of ozone mixing ratio and water vapour from Met. Research Flight information, shown in figure 4.1, we

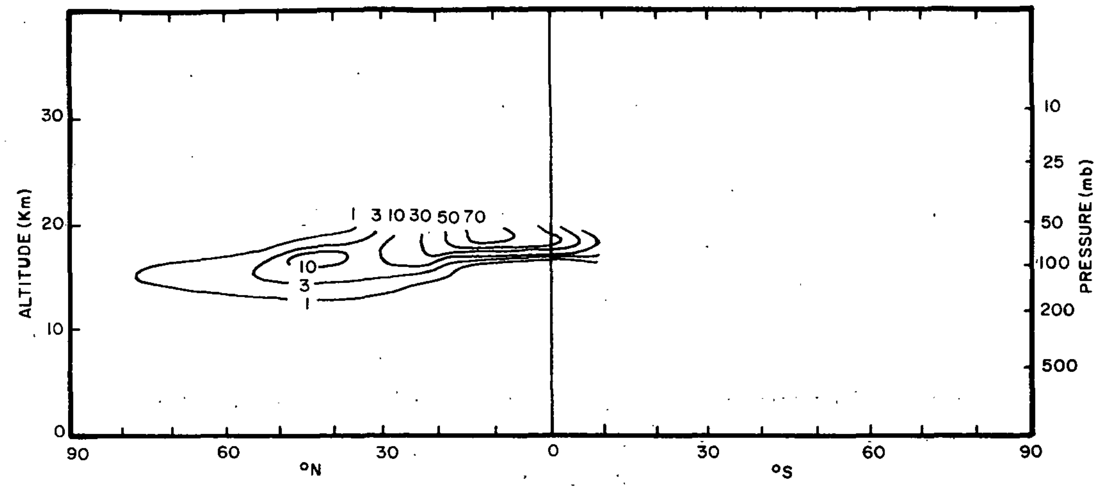

Figure 4. 12.
Meridional Distribution of W^{185} for September-October, 1958 (Newell, 1961)

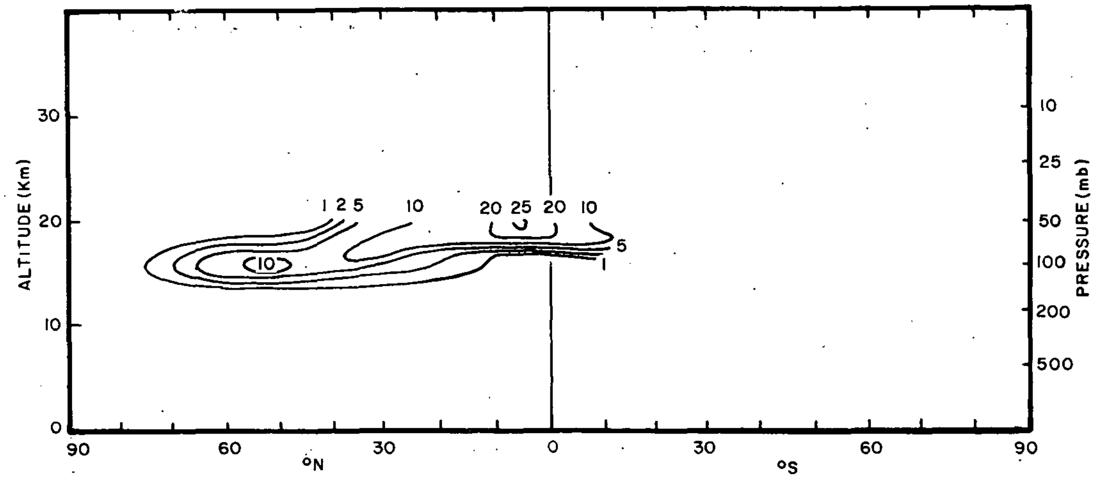

Figure 4. 13.
Meridional Distribution of W^{185} for November-December, 1958 (Newell, 1961)

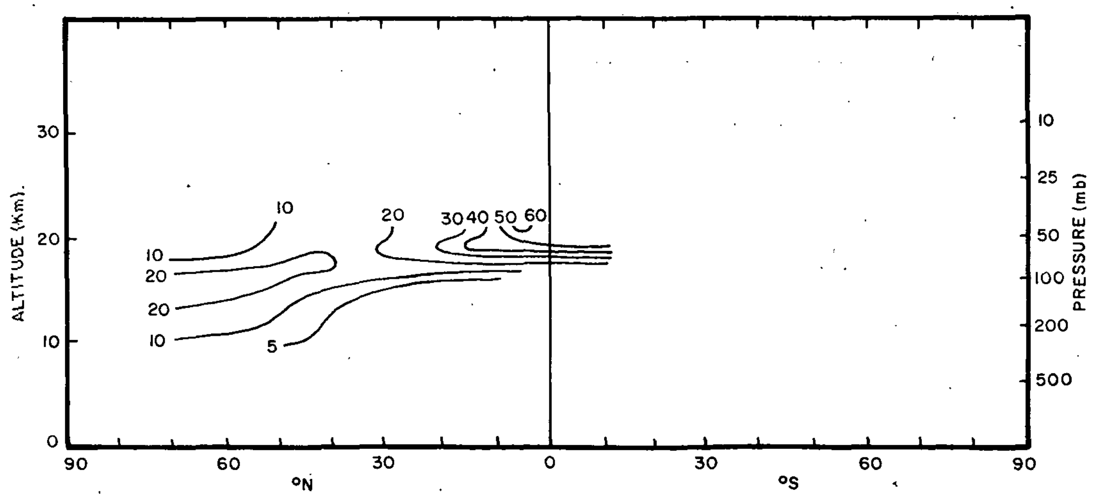

Figure 4. 14.
Meridional Distribution of W^{185} for November-December, 1959
Corrected to 15-8-58 (Feely & Spar, 1960).

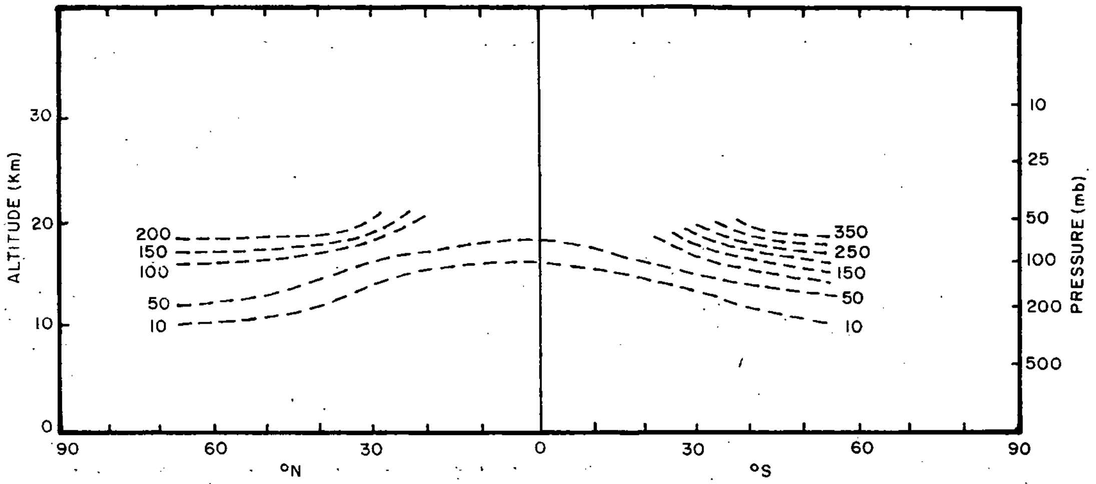

Figure 4. 15.
Meridional Section of Mean Rh^{102} Concentrations in May 1961 from High Level Injection of 'Orange' Test (17° N, Altitude 30 km Burst) Corrected to the Date of the Explosion - 12-8-58.

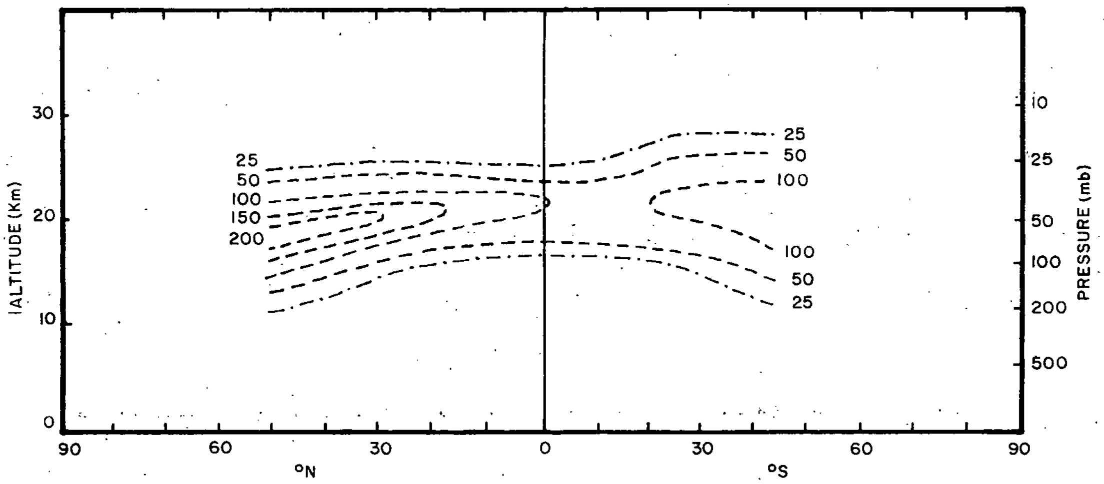

Figure 4. 16.
Meridional Section of Mean Sr^{90} Concentrations for the Period January-June 1957.

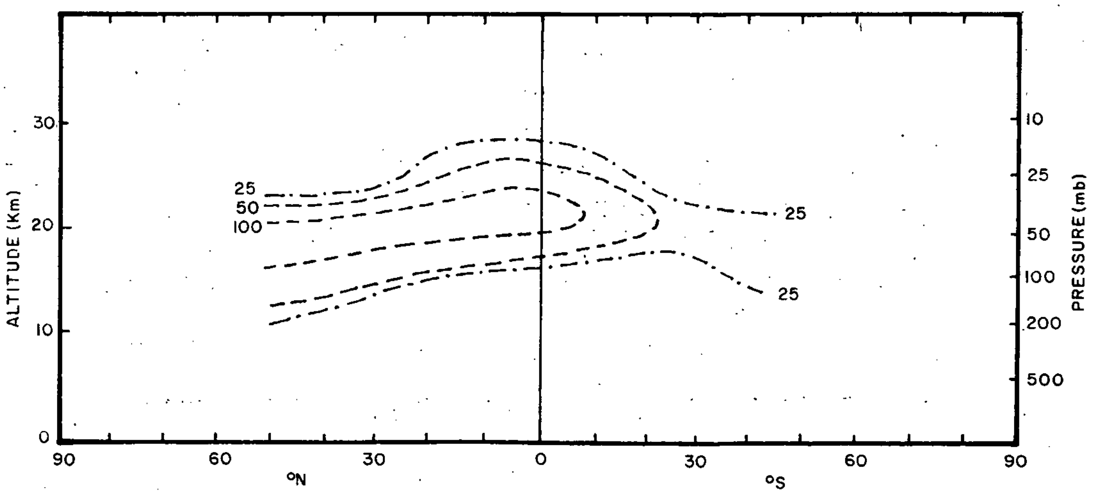

Figure 4. 17.
Meridional Section of Mean Sr^{90} Concentration for the Period January-June 1958.

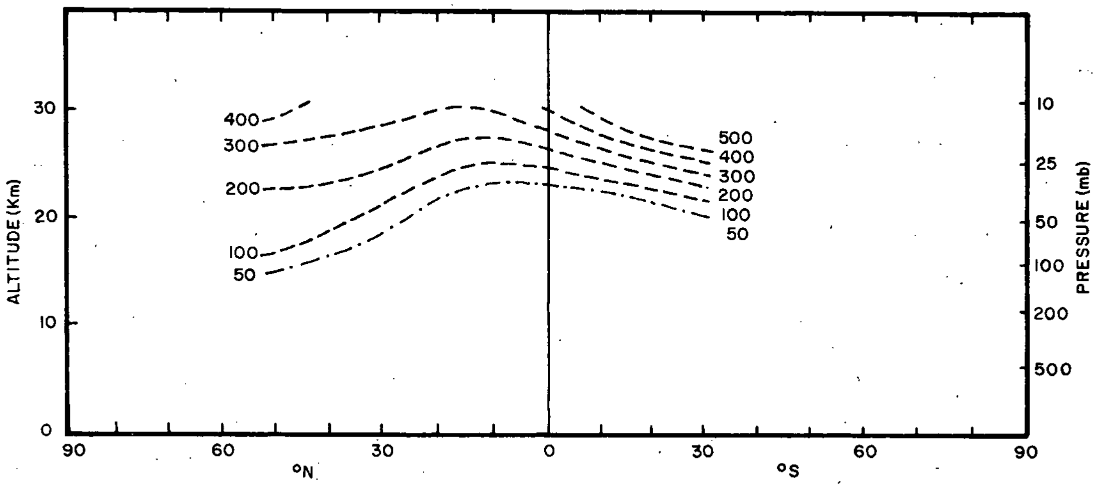

Figure 4. 18.
Meridional Section of 6 Monthly Mean Values of Excess C^{14} for Winter, 1956, Computed from Data of Hageman et al, (1959).

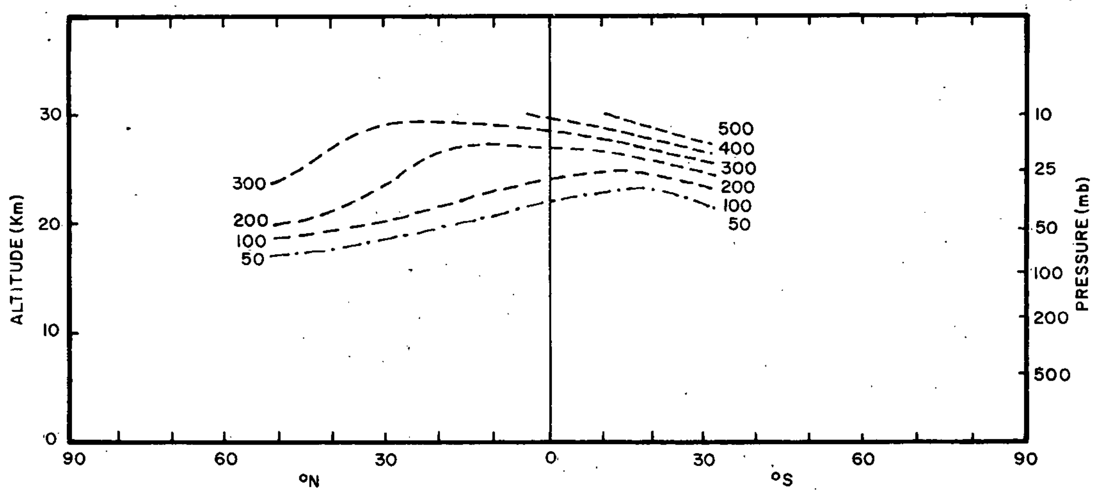

Figure 4. 19.
Meridional Section of 6 Monthly Mean Values of Excess C^{14} for Summer, 1955, from Hageman et al, (1959).

observe that the tongue of ozone rich air with low water vapour content dips down rather markedly in subtropical latitudes from levels normally in the stratosphere to levels more characteristic of tropospheric air. By comparison with the independently drawn mean cross sections of wind and potential temperature in figure 5.1, we observe that the tongue apparently coincides with the region in the vicinity of the wind maximum, where the isentropic surfaces sweep down from the more stable stratosphere into the troposphere. This region may be interpreted as corresponding to the upper tropospheric frontal zone on individual cross sections. Unfortunately, no section for another longitude than that of figure 5.1 is available to support or contradict this apparent relationship.

Analyses relating the total ozone with features of the upper tropospheric and lower stratospheric temperature fields have been carried out by Meetham (1937), Normand (1951, 1953, 1954), Gowan and Leppard (1953) and Johansen (1955) to name a few and the current view may be stated as follows:

1. A high correlation exists between total ozone and both temperature and potential temperature in the lower stratosphere. The correlation drops with the introduction of a 48-hour time lag either way suggesting that the variations result from the same mechanism.

2. The correlation with the tropopause height is significantly lower in winter, during the time of the polar night vortex, than in summer. Ohring and Meunch (1960) found ozone was generally more highly correlated with temperature at 100 mb than 50 mb for European Stations during 1956 and 1957 but the sample was inadequate to detect seasonal variations in the correlation coefficients.

   Normand (1953) took 3 day mean values of ozone at Oxford and temperature in the upper troposphere (300-500 mb thickness) and found a close relation between the deviations from their respective seasonal mean values, but noted some major inconsistencies in winter. Johansen (1955) found similar results for Tromso.

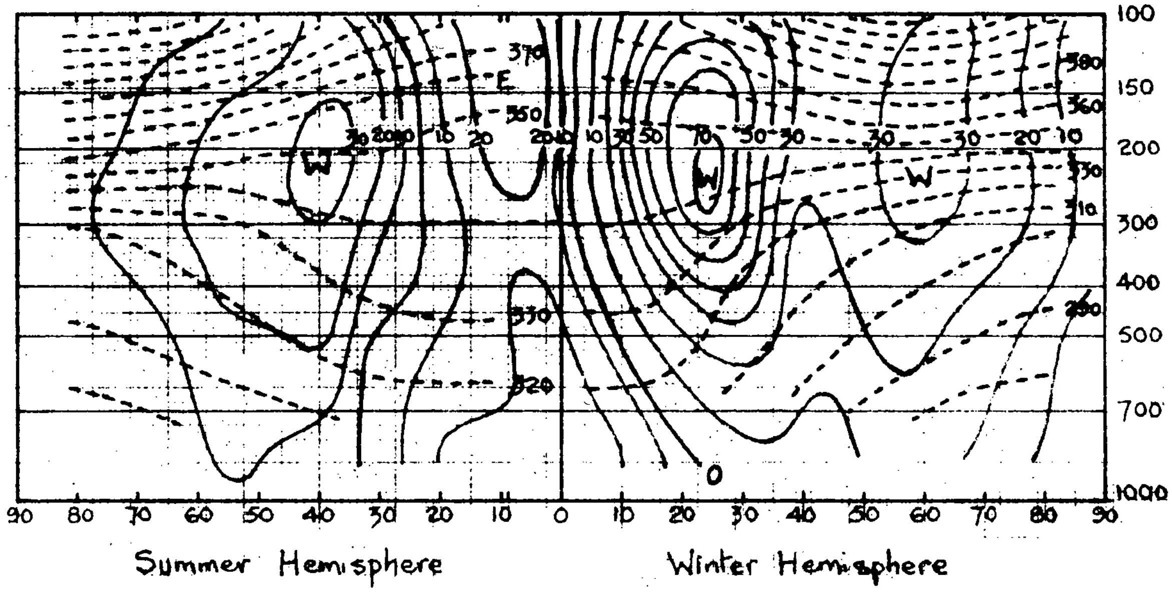

Figure 5. 1.

Mean Sections of Potential Temperature (^{O}K) and Zonal Wind Component (Knots) for O^{O}W.

3. Normand (1951) found total ozone to be most highly correlated with potential temperature at 18 km and Ohring and Meunch (1960), using 100 mb charts ($\sim$ 16 km), found that high total ozone was associated with high temperature, low geopotential height, south winds and cyclonic curvature at that level. The meridional gradient of ozone is generally directed equatorward over the range 35°-65°N so an ozone increase with south wind implies a simultaneous subsidence which overcompensates the negative meridional advection. The correlation coefficient showed little seasonal change.

Ohring and Meunch (1960) concluded from the above analysis and from a study of average departures of total ozone from the spatio temporal mean in relation to the standing eddies, that the total ozone maxima and minima occurred slightly in advance of the long wave troughs and ridges respectively.

The relation between gross β-activity concentration maximum in air and rain with the jet-front complex at 80° W in figure 4.10 suggests transfer of debris from its source may be strongly dependent upon it. We observe a distinct relation between the distribution of S_r^{90} deposition on a hemispheric chart (figure 4.2) after Alexander (1960) and the position of the mean wind maximum from Crutcher (1961). The regions of maximum deposition correspond to the regions of confluence of the polar front and sub-tropical jets to the southeast of the troughs of the middle latitude standing eddies.

While the meridional variations in deposition and in air activity may be related tentatively to the mean position of the jet front complex, the seasonal variation remains to be related to the wind distribution.

It seems plausible to look for similar transfer mechanisms associated with the jet complex of the polar night vortex to those in the troposphere, and it does appear that transfer there is also associated with the westerly jet maximum. The polar stratospheric jet is a winter-spring phenomenon and it is during this period that the lower polar stratosphere shows its highest ozone values of the year. Figures 5.2 to 5.5, obtained by subtracting successive seasonal mean total ozone amounts as shown in figures 4.2-4.5, show a general increase in middle and high latitudes in the 1st and 4th quarters. Moreover, the correspondence between the disproportionately high ozone increase in figure 5.2 and the region of greatest baroclinity in the polar night vortex over arctic Canada in spring (Hare 1960), where short period thermal waves have been observed (Boville et al, 1961) suggests that a major part of the downward transfer may be effected by large scale transient eddy exchange.

5.3. Tracers as related directly to jet front complex

5.3.1. Water vapour

Analyses of Met. Research Flight data on water vapour distributions have been carried out relative to such features as the tropopause by Bannon et al (1957), Murgatroyd et al (1954), Helliwell et al (1956), and Tucker (1957); fronts by Sawyer (1955, 1957), and Miles (1962); and jet streams by Tucker (1957), Murray (1956) and Briggs and Reach (1963).

Vuorela's (1957) analysis of dewpoint depression below 400 mb in selected jet-front complexes over western Europe, suggests that air in the frontal zone is frequently subsiding relative to its environment, and this inference is supported for mid-tropospheric levels by Sawyer's (1957) observation from Met. Research Flight frontal traverse data that the frontal zone is frequently dry relative to its environment.

In the upper troposphere, information from jet traverse flights have been analysed by Murray (1956) and Briggs and Roach (1963) and indicate the existence on many flights of a fold in the mixing ratio isopleths suggesting an intrusion of relatively dry polar stratospheric air into the troposphere below the jet axis in the region corresponding closely to the frontal zone.

On some flights only two traverses were flown resulting in considerable subjectivity in analysis, but on flights like that shown in figure 5.4, the resolution was sufficient that this would not radically alter the pattern.

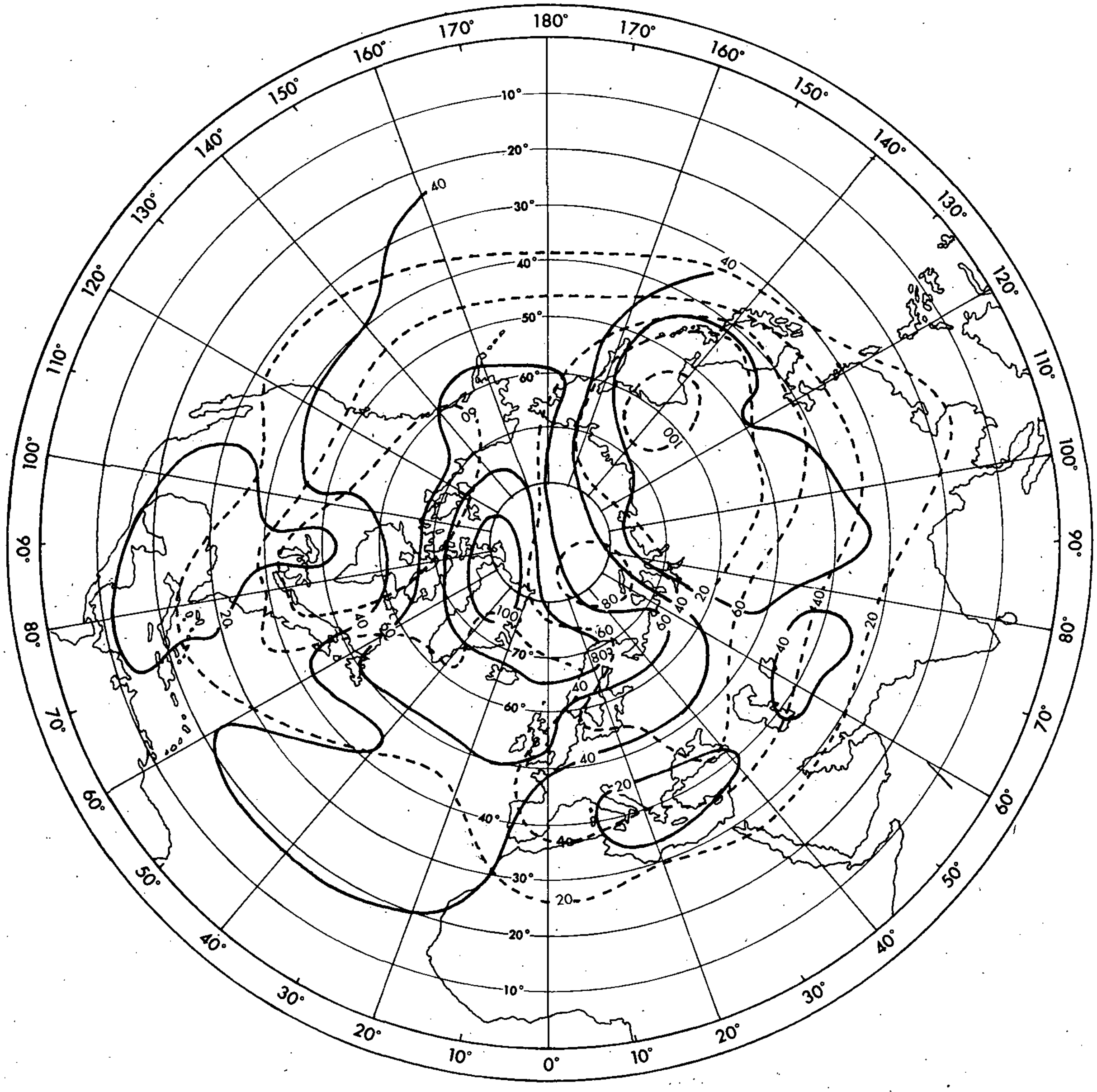

Figure 5. 2.
Isopleths of the Mean Increase in Vertically Integrated Ozone Amounts Over the Northern Hemisphere from Autumn to Winter (Dashed Lines) and from Winter to Spring (Continuous Lines) in cm. atm. S. T. P. 10^{-3}.

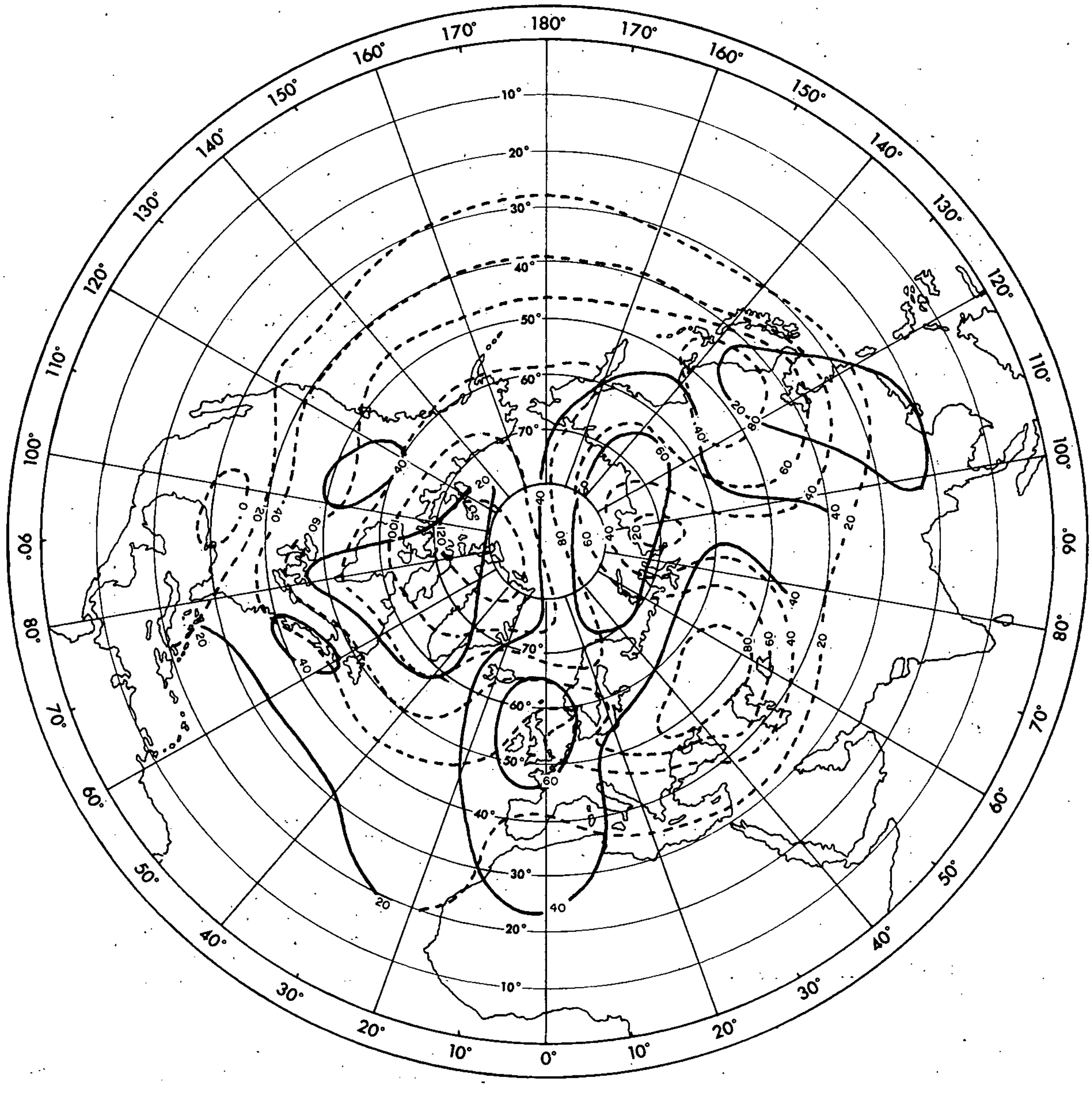

Figure 5. 3.
Isopleths of the Mean Decrease in Vertically Integrated Ozone Amounts Over the Northern Hemisphere from Spring to Summer (Dashed Lines) and from Summer to Autumn (Continuous Lines) in cm. atm. S. T. P. 10^{-3}.

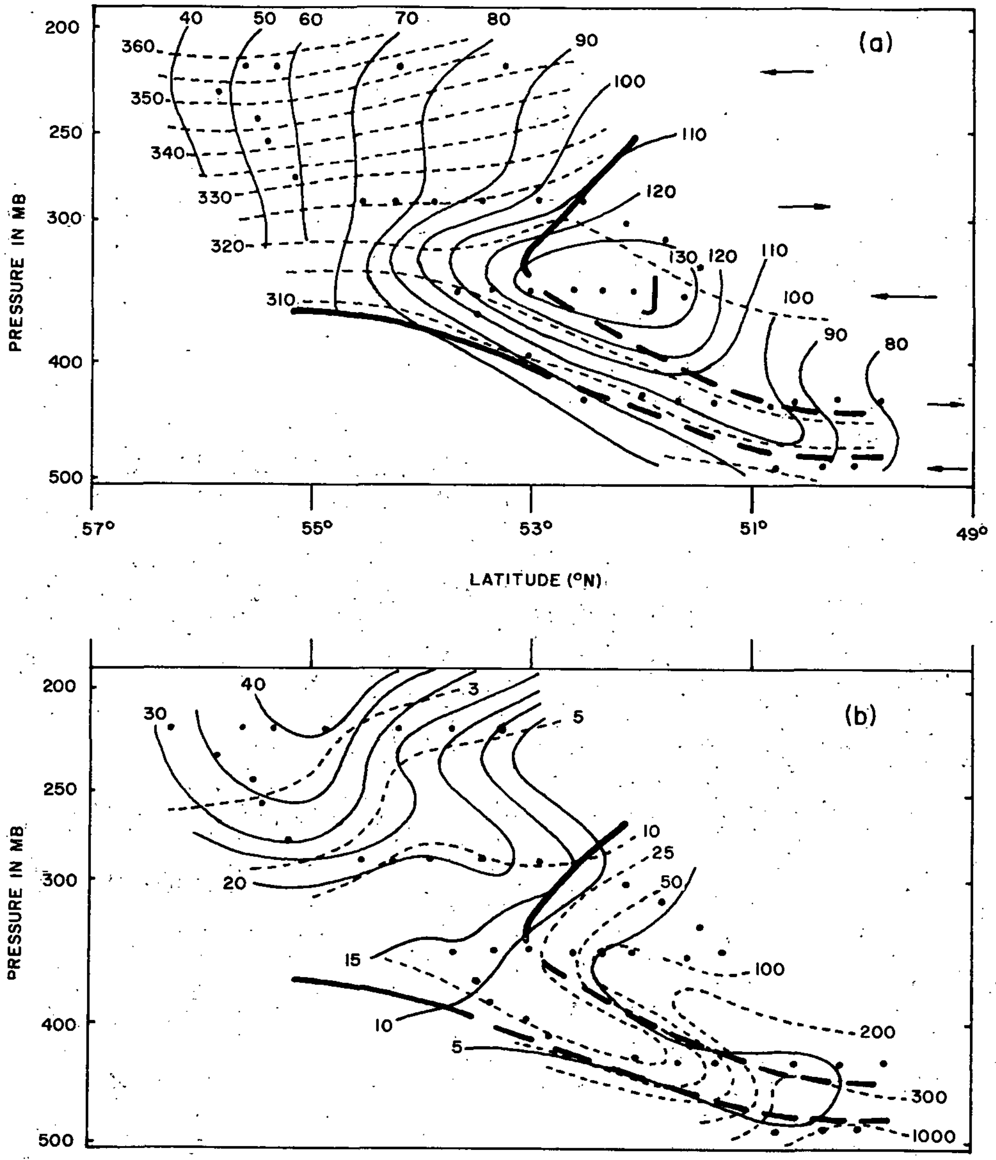

Figure 5. 4.

Analysis of Aircraft Observations on Flight of 8 May 1961. (Briggs & Roach 1963).

(a) ——— Isotachs in knots.

\- - - - - - Potential isotherms in °K.

(b) ——— Ozone mixing ratio in mol. / 10^8 mol.

\- - - - - - Humidity mixing ratio in μ g/g.

The Flight Path is Indicated by the Arrows and the Dots Represent the Points of Observation. The Thick Solid Lines Represent Tropopauses and Frontal Boundaries.

Tucker (1957) inferred a direct transverse circulation about the jet axis in entrance zones and an indirect one in exit zones from an analysis of average frost point departure from the mean value for standard levels above and below the jet axis.

5.3.2. Ozone

Briggs and Roach (1963) analysed the ozone distribution with respect to the jet-front complex for individual cases and statistically, as previously discussed in relation to humidity. The mean distributions show that the intrusion of stratospheric air down the frontal zone below the jet is not a persistent feature of the pattern of transport.

5.3.3. Artificially produced radioactive isotopes

Miyake et al (1960) could detect no systematic variation in the integrated activity 'c' in the tropospheric air column associated with the changes in surface weather conditions but found it to be related to the position of the jet axis and the presence or absence of a marked 500-mb cold trough. The integrated activity 'c' in the rain bearing tropospheric layer is plotted on an abscissa of distance from, and ordinate of pressure of the neighbouring jet axis, as a circle whose area is proportional to 'c' in figures 5.5 and 5.6 for cases with or without a 500-mb trough in the vicinity. The highest values of 'c' are observed to occur when the jet is situated above or a little north of the sampling station and when the jet axis occurs at lower levels.

During the month of March, 1960, an extensive series of observations of S_r^{89}, S_r^{90}, W^{185} and Be^7 concentrations in the upper troposphere and lower stratosphere was made over the U.S.A. Danielsen et al (1962), analysed these observations together with simultaneous values of potential temperature and potential vorticity statistically and by means of case studies in investigating stratospheric-tropospheric exchange processes.

The isopleths of S_r^{90} activity in figures 5.7 and 5.8, drawn consistent with the values plotted over the regions sampled in flight paths normal to the cross sections as indicated by the

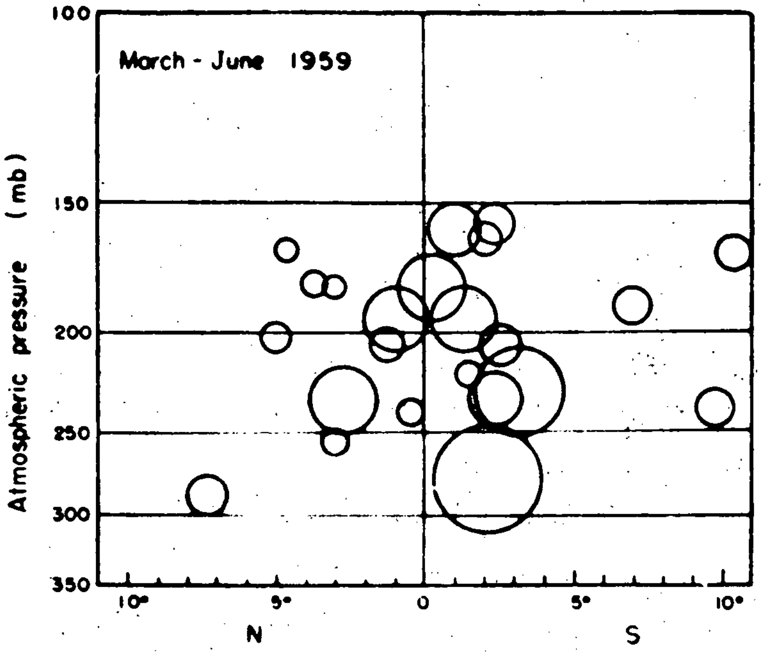

Figure 5. 5.
Air Activity in Rain Bearing Layer and Position of a Jet Stream Accompanied by a Trough at 500 mb.
Surface Area of Each Circle shows Relative Activity.
O Indicates the Position of Observation.

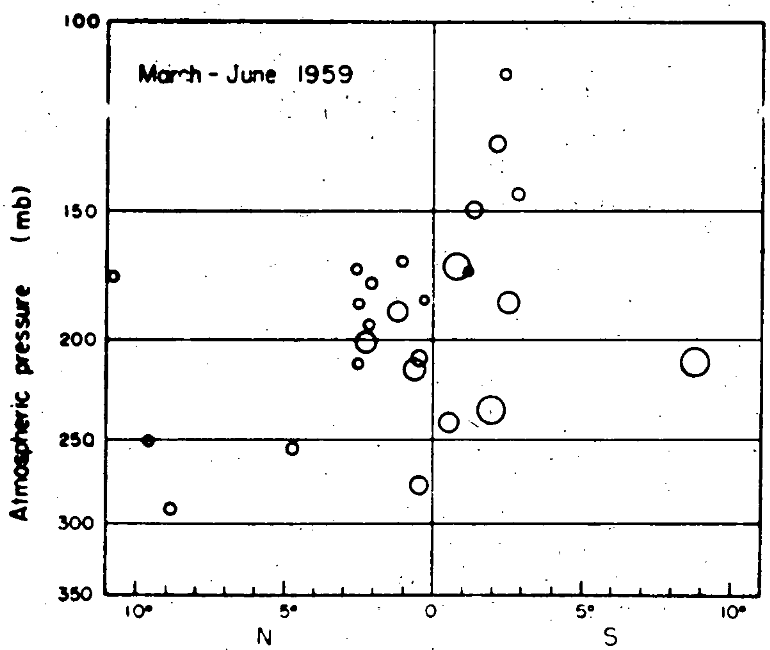

Figure 5. 6.
Air Activity in Rain Bearing Layer and Position of a Jet Stream which Has no Neighbouring Trough at 500 mb.
Surface Area of Each Circle Shows Relative Activity.
O Indicates the Position of Observation.

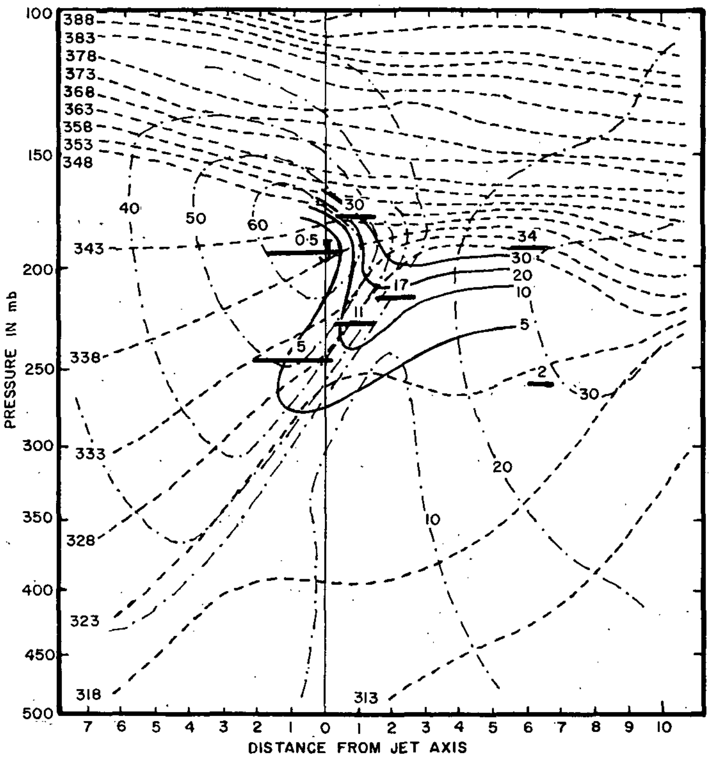

Figure 5.7.
Transverse Section, with Respect to the Jet Axis, of Sr^{90} Concentration in d. p. m. /1000 s. c. f. (Continuous Lines), Potential Temperature in °K, (Dashed Lines), and Isotachs in Knots (Dash-Dot Lines). Heavy Dashes Represent Sampling Traverses. 1 Unit Distance ~ 60 Nautical Miles. (Danielsen et al, 1962).

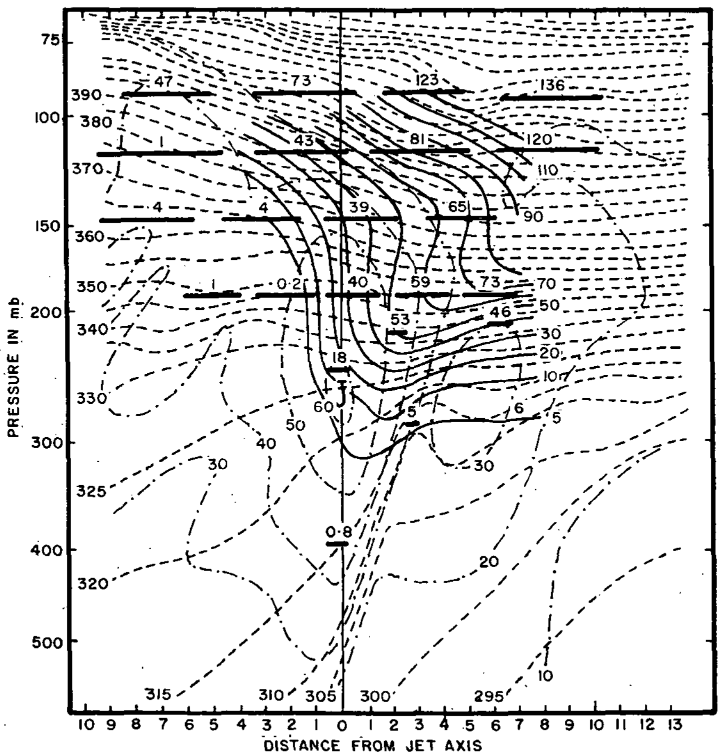

Figure 5.8.
Transverse Section, with Respect to the Jet Axis, of Sr^{90} Concentration in d. p. m. /1000 s. c. f. (Continuous Lines), Potential Temperature in °K, (Dashed Lines), and Isotachs in Knots (Dash-Dot Lines). Heavy Dashes Represent Sampling Traverses. 1 Unit Distance ~60 Nautical Miles. (after Danielsen et al, 1962).

heavy bars, show a tendency for air of stratospheric origin to intrude into the troposphere in the region below the jet on some occasions.

5.5. Case studies and analyses of particle trajectories

Case studies require an accurate four dimensional description of the atmosphere in order that individual parcels may be followed through successive stages of their history and related to the persistent features in analysis, viz fronts, tropopauses and jet streams.

The studies of Reed and Sanders (1953), and Reed (1955), investigating the thermal structure of particular cyclones in the upper troposphere and lower stratosphere at successive stages of development, showed that a pre-existing frontal zone was not prerequisite for cyclogenesis. In fact frontogenesis and cyclogenesis took place simultaneously associated with large horizontal gradients of vertical velocity which tilted the isentropic surfaces in the vertical.

Reed (1955), Danielsen (1959), Staley (1960) and Danielsen et al (1962) showed cases of mass transfer from stratosphere to troposphere within stable layers below the jet, tracing specific air parcels by trajectories based on conservation of potential temperature and potential vorticity over 12-hour intervals. Staley (1960), moreover, noted the existence of exceptionally dry air as measured by standard humidity elements in routine radiosonde ascents, at the final point of those trajectories suggesting subsidence from the lower polar stratosphere.

Staley (1962) interpreted vertical profiles of β-ray activity, made from aircraft observations on days when a dry baroclinic zone either existed or was anticipated within the troposphere, in terms of transfer. Where maxima in the β-ray activity profiles occurred, they were roughly equivalent in height to the stable layer, but maxima were only noted on about half the cases, suggesting the existence of an upper tropospheric frontal zone was not a sufficient condition for subsidence to take place from stratosphere to troposphere. Isentropic trajectories were constructed on those days when observations were suitable to verify the origin of air at the edges and middle of the baroclinic zone. The results showed that in each case of observed high activity in the frontal zone, at least part of the air in the stable layer was of stratospheric origin, whereas in cases showing no activity maximum, the air was found to originate within the troposphere.

6. Transfer as Implied by Tracer Distributions

The tracers discussed may never be considered conservative on a climatological scale unless the correlation coefficients arising in the eddy flux terms are zero. The tracer distributions may be used to advantage however, either to confirm or refute a model or, where possible, to directly evaluate the large scale eddy terms.

Seeking to interpret the observed meridional gradient of total ozone and the absence of diffusive separation in the stratosphere, Dobson (1929) postulated a model of transfer incorporating slow large scale meridional overturning and small scale vertical eddy diffusion. He proposed that air rose through the equatorial tropopause, spread polewards and downwards through the middle and high latitude tropopause, and that vertical eddy mixing took place through the tropopause and lower stratosphere.

Brewer (1949) estimated mean velocities for this circulation from profiles of water vapour in middle latitudes. The balance equation for a conservative property in the middle to high latitude lower stratosphere neglecting horizontal advection may be expressed by:

$$K \frac{\partial^2 s}{\partial Z^2} - \bar{\omega} \frac{\partial s}{\partial Z} = 0 \qquad 6.1.$$

where K is the vertical eddy diffusivity and S the value per unit mass of tracer. Neglecting vertical variation in $\bar{\omega}$ and K and integrating, the equation becomes:

$$\frac{s - s_o}{s_t - s_o} = e^{\frac{\bar{\omega}}{K} Z}$$

where Z is the height measured from the tropopause and s_t and s_o are the values of s at the tropopause and in the upper part of the descending current respectively.

Brewer (1949) applied this equation to humidity profiles in middle latitudes and found reasonable agreement in many cases for a value of $\frac{\overline{\omega}}{K}$ of about -3×10^{-5} cm^{-1}. In the absence of any measured value of K in the lower stratosphere he proposed a reasonable value of 10^{-3} cm^2 sec^{-1}, which implies $\overline{\omega} \doteqdot -3 \times 10^{-2}$ cm sec^{-1} or 25 m day^{-1}. This would result in adiabatic warming of about 0.25°K day^{-1} which might well be compensated by radiative cooling. The complementary heating required in the tropical stratosphere is more difficult to account for though computations of mean annual rate of temperature change by Manabe and Möller (1961) indicate a possibility of slight heating in this region.

The suggestion in section 4 that the stratosphere is almost uniformly dry (.01 - .04 gm kg^{-1}) to an altitude of 30 km throughout the year is consistent with the Brewer-Dobson circulation model.

Interpreting the ozone distribution quantitatively demands a knowledge of the source strength and the rate of leakage of ozone through the tropopause. The latter is unlikely to be greater in summer than in winter, nor is there much seasonal variation in the source strength and distribution, so differences in seasonal distributions are likely to be due to variations in transfers. The observed spring build-up of ozone in the winter lower stratosphere is thus consistent with maximum circulation strength in winter and spring, and little or no circulation in summer. The generally lower ozone mixing ratios in the summer lower stratosphere (figure 4.7) in spite of the seasonal increase of source strength at a given level could be accounted for by the poleward arm of the meridional cell being confined to a lower level during this season, and consequently the descending current being relatively poor in ozone. Moreover, the decrease in slope of the ozone isopleths in summer (figure 4.7) implies a weakening of the circulation.

The low ozone concentration in the tropical lower stratosphere (Ramanathan and Kulkarni, 1960) in all seasons, which are less than would be expected from photochemical equilibrium (Dütsch, 1956) strongly suggest a slow ascending current.

The obvious features of the analysis of radioisotopes may be summarised as follows:

1. Low concentrations in the lower stratosphere relative to the tropopause of S_r^{90}, W^{185}, Rh^{102} and excess C^{14} are observed in low latitudes in figures 4.12-4.19 consistent with the slow ascending current of the Brewer-Dobson model.

2. Figures 4.18-4.19 from Hageman et al (1959) show the tropical lower stratosphere to be poorer in excess C^{14} in winter than summer, in each hemisphere, implying that the mean ascent and consequently the meridional cell is stronger in winter than summer.

3. The axis of maximum concentration of S_r^{90} in figures 4.16 and 4.17 slopes downward from the equator towards the poles in both hemispheres, and shows little vertical displacement with time, in spite of being injected by all nuclear bursts and consequently having a highly variable source. This is apparently inconsistent with the Brewer circulation since the ascending and descending currents of the meridional cell would be expected to advect the levels of maximum S_r^{90} along with them.

4. The axis of maximum concentration of W^{185} in figures 4.12-4.14, like slopes downward from the equator towards the poles in both hemispheres and shows little vertical displacement with time. Moreover, transport, neglecting large scale eddy fluxes fails to account for the meridional spread of the W^{185}, the lack of meridional move-

ment of the concentration maximum, and the lack of subsidence in the middle latitude level of concentration maximum over the period considered.

Stebbins (1960) from analysis of the rate of change of vertical profiles of W^{185}, estimated values of the vertical eddy diffusivity of 10^3 cm^2 sec^{-1} in the tropical stratosphere, and 4×10^4 cm^2 sec^{-1} in the tropical stratosphere, and 4×10^4 cm^2 sec^{-1} in middle latitudes. This latter value is over an order of magnitude greater than the value assumed by Brewer (1949) and would result in a much faster meridional cell.

We now invoke the concept of large scale eddy mixing in an attempt to explain the inconsistencies. Molla and Loisel (1962) computed zonally averaged covariances of vertical and meridional wind components in the lower stratosphere and showed they implied a mixing angle (i.e. the average angle at which transport by an isotropic mixing by slantwise convection occurs) close to that of the mean isentropic surfaces. No such direct evidence of eddying motion exists in the middle stratosphere, but by analogy with the tropospheric systems, transfer by waves associated with the polar night vortex seems plausible. The seasonal mean distributions of ozone in section 5.2 were interpreted as evidence of eddy transfer by transient eddies.

The Brewer-Dobson model of stratospheric circulation is modified in figure 6.1 to incorporate a pattern of large scale eddy mixing consistent with the preceding tentative data interpretation.

The meridional sections of ozone mixing ratio in figure 4.7 are also consistent with the pattern of eddy mixing shown in figure 6.1. The generally lower values observed in the summer lower stratosphere might then be partially accounted for by the lower levels to which the stratospheric eddy mixing was confined in the absence of the polar night jet.

Newell (1961) has estimated the horizontal flux of ozone by transient eddies in the lower stratosphere. In the absence of a sufficient number of vertical profiles of ozone, he assumed the total ozone anomaly to approximate that in the lower stratosphere

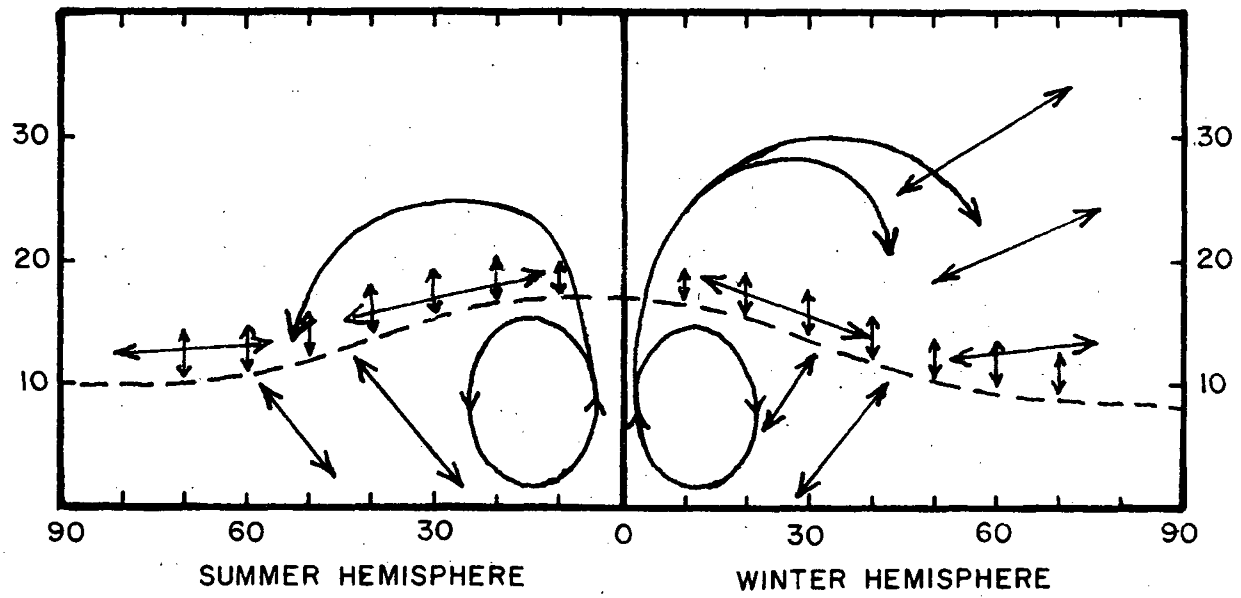

Figure 6. 1.
The Brewer-Dobson Model of Statospheric Circulation Incorporating, In Addition, Macro-Eddy Mixing. The Short Arrows Represent Vertical Eddy Diffusion, and the Long Oblique Arrows, Macro-Eddy Mixing.

(12-24 km). Newell estimated the horizontal eddy flux from 25 stations, for three monthly periods throughout the I. G. Y. using for alternative estimates, the winds at the 50 and 100 mb levels. In middle latitudes he found the eddy flux directed polewards for both estimates with strongest flux in the 1st and 4th quarters, but for three stations north of 60°N where the stratospheric isentropes reversed their slope in winter, it was found to be southward. These results are consistent with the proposed eddy transfer pattern.

Newell (1961) attempted to evaluate the relative contributions of the various transfers to the lower stratospheric spring build up of ozone in high latitudes, examining the ozone budget north of 55°N. The evaluation of both mean meridional transport $(\overline{O_3})(\overline{V})$ and that by standing eddies $(\overline{O_3 V}) - (\overline{O_3})(\overline{V})$ require an accurate evaluation of mean meridional velocity and mean ozone. The sample used by Newell is clearly too small to be representative and so the results can at best only represent a rough approximation, and might even be incorrect in sign. He estimates a zonal temporal mean meridional velocity for the 100-25 mb layer, directed poleward, of 6 cm sec^{-1} during the first quarter. His reasons for adopting such a value are unconvincing, (he takes unpublished estimates of the wind in this layer in summer by Barnes which are directed equatorward, reverses the direction, but keeps the same magnitude). Since this value is consistent with an upward extrapolation of the value at 100 mb of Palmén and Vurela (1963) and the values computed by Murgatroyd and Singleton (1961) as necessary to maintain balance between the radiative sources and sinks, it may not be unreasonable. On the further assumption that a third of the total ozone in a vertical column is involved in the lower stratospheric transport processes, Newell estimates the transport across 50°N due to transient eddies mean advection and standing eddies to be 9.0×10^9, 1.6×10^9, and 2.5×10^9 atm cm cm^{-2} sec^{-1} respectively.

Finally, the introduction of the scheme of large scale eddy mixing helps to explain the distribution of certain radioisotopes. The W^{185} debris which was injected at 12°N in summer 1958 appeared to spread polewards and downwards in a manner consistent with eddy mixing along a slope somewhat greater than that of the mean isentropes, implying the eddy motion to be energy consuming.

A model of the physical processes contributing to transfer of air between stratosphere and troposphere may also be inferred from the data in section 5.

The relation between the westerly wind maximum and the deposition of radiodebris concentrations in figures 4.2 and 4.10 suggested that stratospheric-tropospheric transfer was associated with the jet and the distribution of integrated tropospheric radiodebris of Miyake et al (1960) in figure 5.5 and 5.6 which exhibit maximum to the right of the axis.

Sporadic intrusions of stratospheric air from the lower polar stratosphere into the troposphere in the vicinity of the frontal zone below the jet axis have been inferred from humidity and ozone data by Briggs and Roach (1963), figure 5.6; from S_r^{90} by Danielsen et al (1962) - figures 5.9 and 5.10 and from β activity maxima on vertical profiles by an isentropic trajectory technique (Staley, 1962). These are clearly inconsistent with transfers by a mean indirect circulation as implied by the analysis of mean component of the wind normal to the jet as computed by Murray and Daniels (1953). Tucker's (1957) analysis of humidity in relation to the jet, however, suggests that indirect cells alternate with indirect about the jet and the combined evidence is consistent with these intrusions taking place in a direct circulation in entrance zones. A simple dynamical analysis by Sawyer (1958) supports this inference.

The examples given have illustrated the power of tracers and the rich rewards in understanding the physical processes within the atmosphere to be expected with improvement in observational techniques.

APPROVED,

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