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The Sources and Sinks of Water Vapor in the Stratosphere

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BOULDER, COLO.
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CONTENTS

THE SOURCES AND SINKS OF WATER VAPOR
IN THE STRATOSPHERE

Helmut K. Weickmann and Charles C. Van Valin

The average water vapor concentration in the lower stratosphere is about 2.5 ppmm according to the most reliable measurements. There is an average annual north-to-south gradient of less than 0.5 ppmm, a maximum mixing ratio over the Intertropical Convergence Zone (ITCZ) of from 3 to 4.5 ppmm, and an increase of about 50 percent in the mixing ratio with increasing altitude up to the stratopause. The mother-of-pearl clouds occasionally observed in high latitudes are evidence that higher mixing ratios sometimes occur.

Water vapor enters the stratosphere by three primary mechanisms: (1) Hadley cell updrafts, including mean rising motions of large-scale air masses, as well as severe convective storm penetrations of the tropopause over tropical regions; (2) severe convective storm penetrations of the tropopause in the midlatitudes during the warmer months; and (3) oxidation of methane introduced from the troposphere. Volcanoes are occasional minor sources of water vapor. Exchange of tropospheric and stratospheric air along the jet stream systems and tropopause breaks could represent a very large source of moisture. However, it is likely that in most cases the moisture is efficiently returned to the troposphere by poleward and downward motions.

Removal of stratospheric moisture during the polar night by the formation of ice particles is the only sink mechanism which is specific for water vapor. The north-to-south gradient in stratospheric water vapor is due to the lower temperatures and the resulting greater effectiveness of the Antarctic sink.

1. INTRODUCTION

1.1 Present State of H_2O Concentration in Stratosphere

To date, Mastenbrook (1971, 1974) has conducted the most extensive series of measurements of stratospheric water vapor at a single location. At Washington, D.C., he found a trend of increasing mixing ratio from a mean of about 2 to about 3 ppmm (variability, 1 to 5 ppmm) from 1963 through 1969 or 1970, but the mixing ratio declined in 1971 and has generally remained below 3 ppmm since then (fig. 1). Sissenwine et al. (1968) found mixing ratios in the lower stratosphere at Chico, Calif., to be typically about 3 ppmm at the minimum and increasing with altitude to about 25 km. The mixing ratio reached a maximum of from 10 to 40 ppmm within a few kilometers of 25 km.

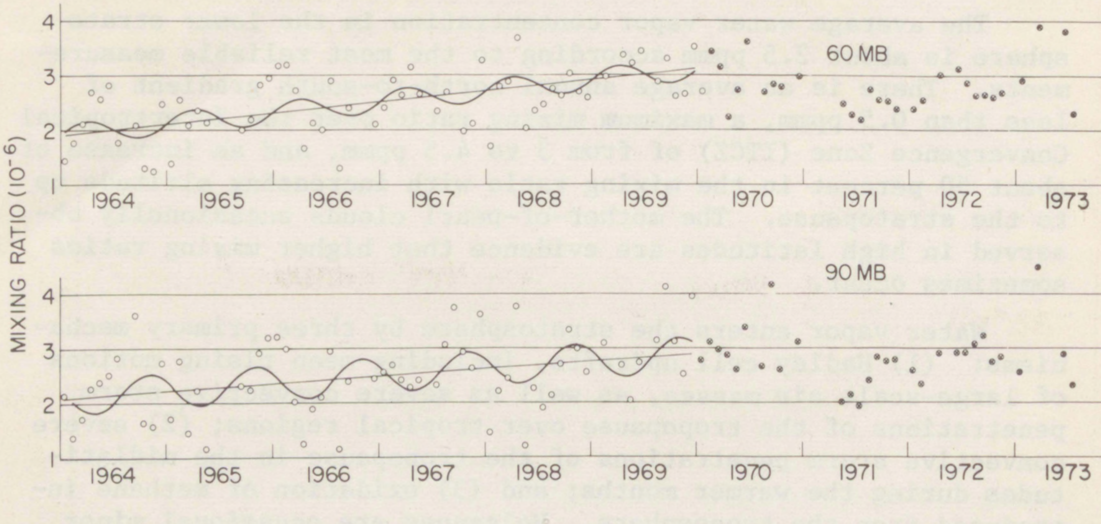

Figure 1. Mixing ratio data for two selected pressure levels over Washington, D.C. Each circle represents one observation. The curves represent the best fit of a linear trend and annual cycle to the first 6 years of data. Cross-marked circles are data collected after the time-series analysis (Mastenbrook, 1974).

McKinnon and Morewood (1970) deduced water vapor concentrations from solar spectra recorded from an aircraft flying at 17.7 km along a generally north-to-south track. During the 1967-68 Northern Hemisphere winter, they found a maximum concentration of 1.75 ppmm above the flight track at 65°N and a progressive decline to a minimum of 1.25 ppmm near 30°S. Most measurements were done in the vicinity of Albuquerque, N. Mex., and the data, collected over a period of 13 months, show a seasonal change of about 17 percent. However, McKinnon and Morewood found the maximum mixing ratio in January, with the minimum in June, contrary to the findings of Mastenbrook (1971). Recently, Kuhn et al. (to be published) measured water vapor above the aircraft during 12 meridional flights of a WB-57F aircraft at altitudes of 15.2 to 19.7 km. These flights, made during September, October, and November 1973, and January 1974, traversed latitudinally overlapping segments between latitudes 75°N and 52°S. Figure 2 is a preliminary presentation of all data collected on these flights. Observations were made at the rate of one per second; raw data points are the result of 1-minute averages. On the assumption of a constant mixing

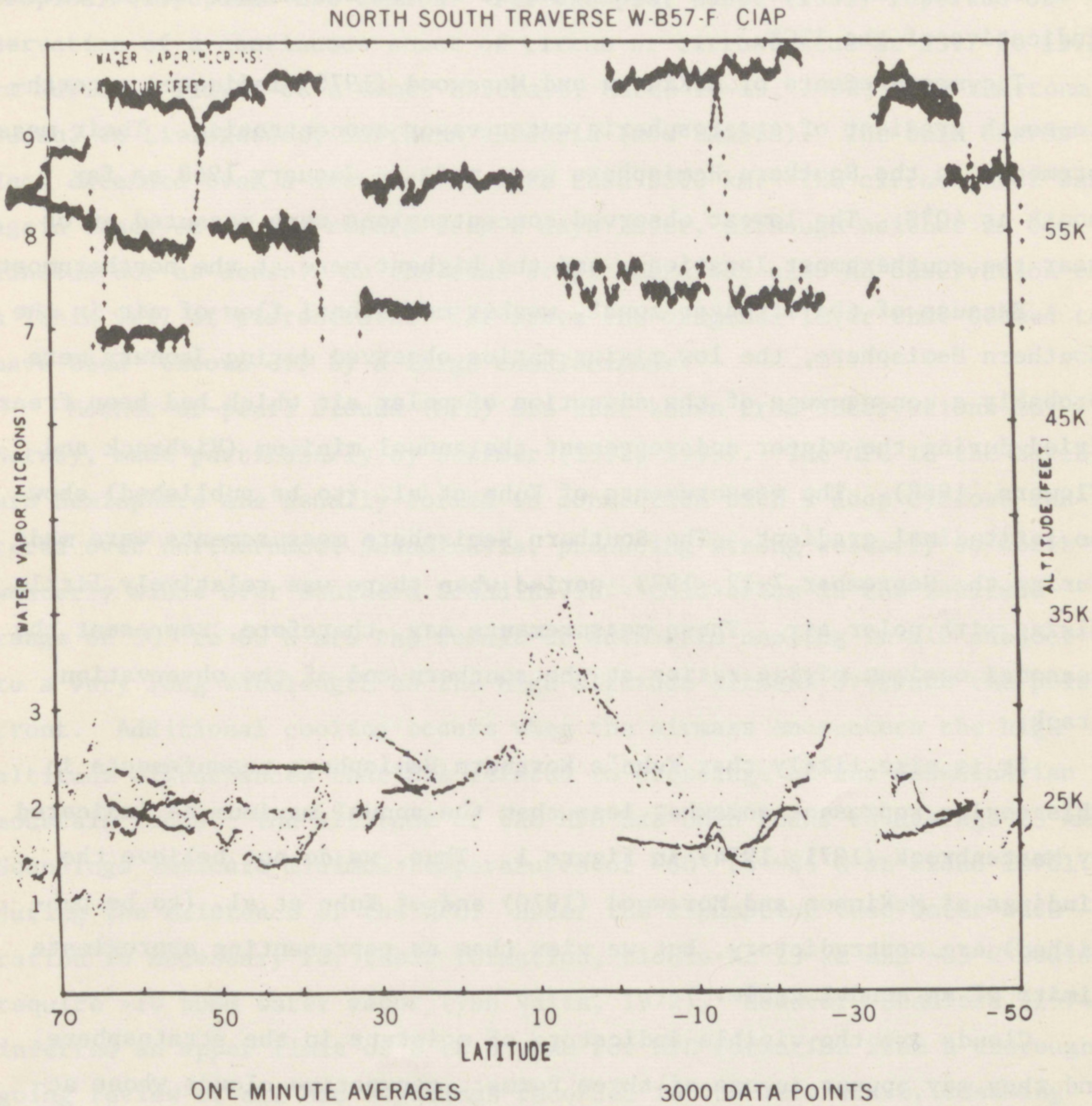

Figure 2. Water vapor density above the flight altitude during Sept., Oct., and Nov. 1973, and Jan. 1974. Positive designation for latitude indicates the Northern Hemisphere; negative designation indicates the Southern Hemisphere (Kuhn et al., to be published).

ratio above the aircraft, 1 μm of water vapor is equivalent to 0.9 ppmm at 15.2 km (50,000 ft), 1.2 ppmm at 17.5 km (57,500 ft), and 1.9 ppmm at 19.7 km (65,000 ft). Significant features seen in this preliminary presentation are mixing ratios over the Intertropical Convergence Zone (ITCZ) from 3 to 4.5 ppmm, midlatitude mixing ratios mostly in the range from 2 to 3 ppmm, and a latitude span of about 20° in the higher mixing ratios indicative of the ITCZ.

The measurements of McKinnon and Morewood (1970) indicated a north-to-south gradient of stratospheric water vapor concentration. Their measurements in the Southern Hemisphere were made in January 1968 as far south as 40°S. The lowest observed concentrations were measured at or near the southernmost locations, and the highest were at the northernmost.

Because of the stronger zonal, weaker meridional flow of air in the Southern Hemisphere, the low mixing ratios observed during January were probably a consequence of the advection of polar air which had been freeze-dried during the winter and represent the annual minimum (Viebrock and Flowers, 1968). The measurements of Kuhn et al. (to be published) show no latitudinal gradient. The Southern Hemisphere measurements were made during the September 7-12, 1973, period when there was relatively little mixing with polar air. These measurements may, therefore, represent the seasonal maximum mixing ratios at the southern end of the observation track.

It is also likely that Kuhn's Northern Hemisphere measurements in this series represent somewhat less than the annual maximum as indicated by Mastenbrook (1971, 1974) in figure 1. Thus, we do not believe the findings of McKinnon and Morewood (1970) and of Kuhn et al. (to be published) are contradictory, but we view them as representing approximate limits of an annual cycle.

Clouds are the visible indicators of moisture in the stratosphere, and they may appear in one of three forms: convective clouds whose active turrets rise past the tropopause; cirrus clouds which form above the tropopause, probably as a result of convective activity; and mother-of-pearl clouds that are occasionally observed in the high latitudes during the winter at altitudes from 17 to 31 km. In addition, one should include aircraft contrails because they indicate layers of high humidity, that is, ice supersaturation in the layers where they form.

There are occasional reports of cirrus clouds in the stratosphere, but they are essentially informal oral communications rather than written reports in the published literature. Extended cirrus layers near the tropical tropopause are common. For example, Durst (1953) reported observation of a continuous sheet of cirrus or cirrostratus at 13.7 to 15.2 km during a flight of a Comet aircraft, March 18-19, 1952, from Khartoum, Sudan, to Livingstone, Northern Rhodesia (now Zambia). The thin cirrus deck extended over a distance of more than 3000 km. The cirrus sheet was again observed on the return trip 2 days later, although neither as continuous nor as dense. On the same trip, there was also an observation of a "thin veil of cirrostratus" far above the original layer that seemed to have been "thrown off by a large cumulonimbus."

Mother-of-pearl clouds (MPC) are best known from observations in Norway, made particularly by Størmer (1932, 1939). The MPC in the Northern Hemisphere are usually formed in connection with a deep cyclone centered over northernmost Scandinavia, producing strong westerly to northwesterly winds over southern Scandinavia. Cold areas in the latitude range of 50° to 60°N are the result of adiabatic cooling in air subject to a very long wavelength as the high altitude airmass overruns the polar front. Additional cooling occurs when the airmass encounters the high altitude disturbances that are related to crossings of the Scandinavian mountain range. The altitude of the MPC has been found to average 23 km. Soundings indicate minimum temperatures of -80° to -85°C at cloud level during the existence of the MPC. Under the assumption that water saturation is necessary for their formation, clouds at 23 km and -85°C would require >10 ppmm water vapor (Van Valin, 1972). However, Stanford (1974) inferred an upper limit of 6 to 8 ppmm for MPC formation from a thoroughgoing review of all MPC sightings recorded in the literature, assuming that the clouds form at ice saturation.

The most detailed observations of MPC have been accomplished by Størmer (1932). He has described a cloud which formed on January 13, 1929, at 25-km altitude, showing characteristics of color that can best be explained by assuming that it formed as a water cloud at temperatures above -40°C. The cloud had a brightly shining, iridescent head with colors parallel to the edges and with a long tail of the bluish-gray color seen in cirrus clouds during the evening. We have observed such cloud types in connection with mountain waves over the Alps and the Rocky Mountains. In clear air, the clouds form initially as water clouds but convert by freezing to ice clouds. Because -40°C is usually accepted as the threshold temperature for the homogeneous freezing of water, it could be argued that the MPC described by Størmer would have to be formed at that or at a warmer temperature. If this were the case, the mixing ratio required would be as large as 2775 ppmm.

A series of interesting artificial clouds that formed following the burst of pilot balloons (pibals) was observed and described by Kuhlbrodt (1931) during the German Atlantic "Meteor" Expedition of 1925-27. Pibal ascents made over the Atlantic Ocean between 11°N and 9.5°S frequently produced, upon bursting of the balloon, a small cirrus cloud of diameter about 50 m which was observable for 1/4 to 1/2 min. The most frequently occurring level of burst was at 18 to 19 km. From the wind distribution, Kuhlbrodt concluded that the tropopause was at 16½ to 17 km. This indicated the existence of humidities close to ice saturation at elevations as much as 2 km above the tropopause, resulting from the upward flux of water vapor in the ITCZ. Figure 3 (Kuhlbrodt) gives the locations of the pibals within the tropical zone which did (+) or did not (-) develop a cirrus cloud. At no other locations during the very extended expedition were similar clouds observed.

1.2 Residence Time

Because the exchange time varies with location, there is no unique "residence time" applicable to all parts of the stratosphere; it will be unique for each parcel of the stratosphere, depending upon latitude, altitude, and atmospheric conditions. Although the lower stratosphere is strongly coupled to the upper troposphere and has an exchange half-time of a year or less, the length of time required for exchange of upper

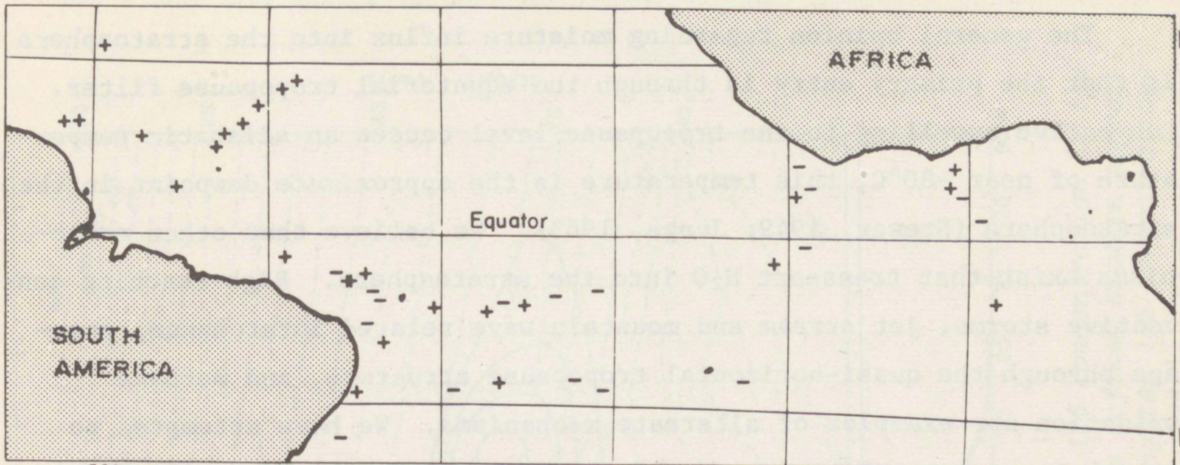

Figure 3. Distribution of cases with (+) and without (-) cirrus cloud formed after pibal burst (Kuhlbrodt, 1931).

stratospheric parcels with those in the troposphere is drastically longer. Nevertheless, the term "residence time" is useful when applied to specific injections, such as nuclear detonations, convective storm penetrations, or aircraft engine effluents.

Residence half-time of nuclear debris in the lower stratosphere is found between 6 and 15 months (Machta and Telegadas, 1973; Machta et al., 1970; Karol, 1970). Mass removal is at a maximum in late spring, involving an annual variation in hemispheric mass of 20 percent. An estimated 70 percent of the stratospheric mass is removed each year, equivalent to an annual removal of all air between the tropopause and about 20.5 km.

Air parcels in the lower stratosphere are transported by a variety of wave motions and by zonal and meridional circulations. Most wave motions have their origin in the troposphere and are gradually damped as they move upward in the stratosphere. Above about 20 km, these transient eddies forced from the troposphere are completely damped out and are replaced by standing eddies. Eddies are considered to be the principal agent for mixing in the stratosphere.

2. SOURCES OF STRATOSPHERIC MOISTURE

The general opinion regarding moisture influx into the stratosphere is that the primary entry is through the equatorial tropopause filter. Convective upwelling to the tropopause level causes an adiabatic temperature of near -80°C; this temperature is the approximate dewpoint in the stratosphere (Brewer, 1949; Junge, 1963). We believe that other mechanisms exist that transport H_2O into the stratosphere. High-reaching convective storms, jet stream and mountain wave related interchange, seepage through the quasi-horizontal tropopause structure, and methane oxidation are examples of alternate mechanisms. We have attempted to attach transport values to some of these processes which we will discuss subsequently.

2.1 Hadley Cell Circulation

The powerful circulation process of the northern and southern hemispheric Hadley cells is related to the ITCZ and its meridional movements during the seasons. Their circulation mechanism reaches above the tropopause at least to the 10-mb level (Vincent, 1968).

Beginning on May 13, 1958, and continuing into the summer of 1958, several moderate-yield nuclear devices were detonated approximately at tropopause level in the equatorial Pacific (11°N). The upper part of figure 4 (after Newell, 1962) represents the Tungsten 185 concentration in the atmosphere during September and October 1958. The tropopause level is indicated in the figure between approximately latitudes 40°N and 40°S. The lower part of figure 4 summarizes Tungsten 185 concentrations at the surface along longitude 80°W in July 1958. From this, it appears that the descending parts of the Hadley circulation are responsible for the bulk of the radioactive deposition. The surface air chart for July 1958 was used because it contains several more data points than do the September and November 1958 charts, but is not otherwise different. It is noteworthy that only a few weeks passed between the injection of the Tungsten 185 and its discharge at the surface.

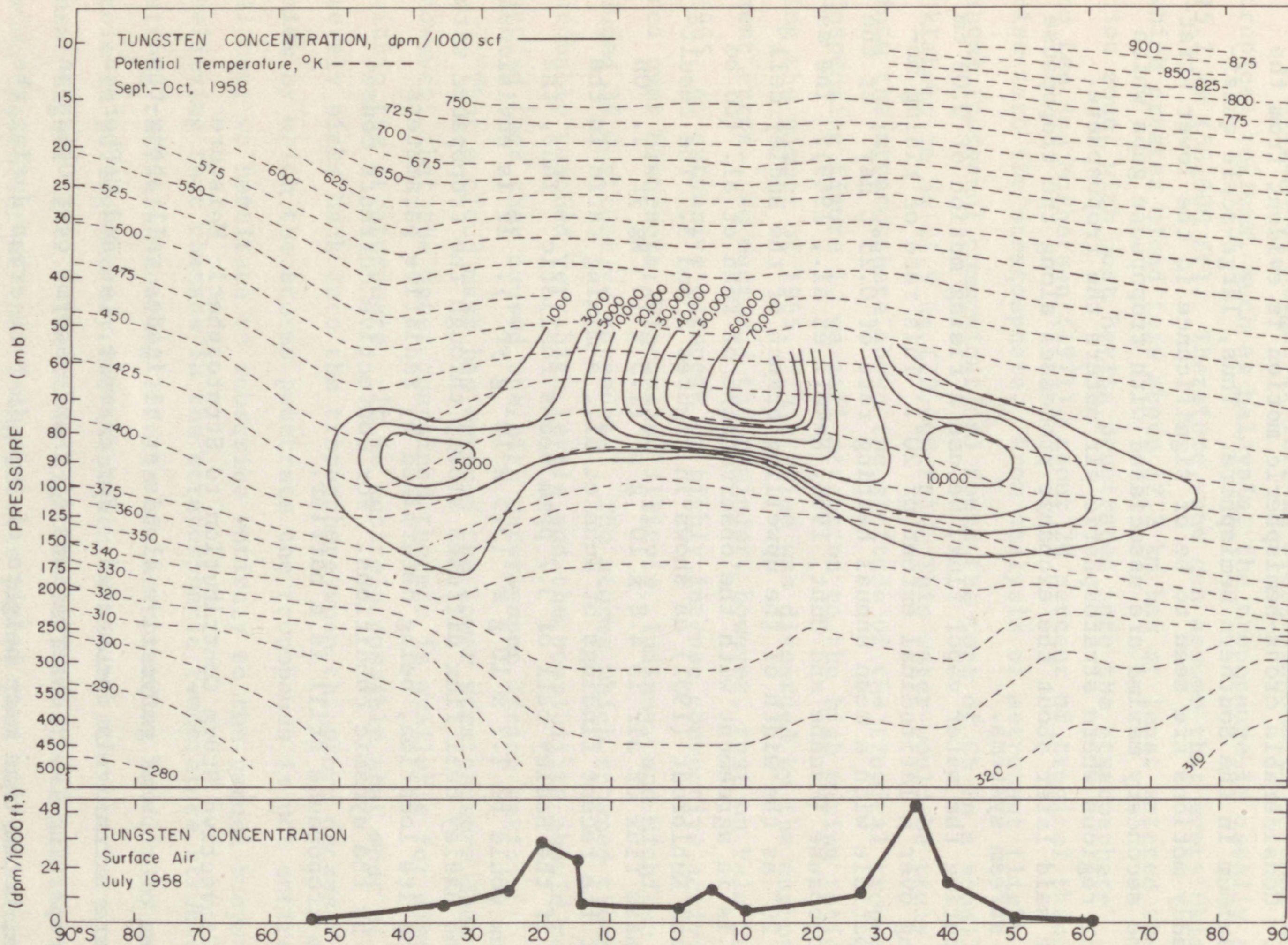

Figure 4. The upper part of this figure represents the Tungsten 185 concentration in the atmosphere during Sept. and Oct. 1958. The tropopause level is defined between approximately latitudes 40°N and 40°S. The lower part summarizes Tungsten 185 concentrations at the surface along longitude 80°W in July 1958 (after Newell, 1962).

The Tungsten 185 was injected into the upper troposphere and lower stratosphere within the ITCZ. Into the fall and winter (Northern Hemisphere), the zone of maximum concentration moved southward with movement of the ITCZ. Considerable cross-hemispheric motion is evident from the secondary maximum in the Southern Hemisphere. Thus, large-scale quasi-horizontal eddy motions are seen to be of significance in the lower stratosphere. The secondary maxima are associated with tropopause gaps which are important regions where stratospheric air enters the troposphere. More will be said later about the exchange processes along the tropopause gap and jet stream systems.

In computing the water vapor flux, we use a rising motion of 0.03 cm s^{-1} operating over a latitudinal extent of 20°. This rate of rising motion is compatible with a mean annual heating rate of 0.25°C day^{-1} at the tropical tropopause (Manabe and Hunt, 1968; Newell et al., 1969). The assumption of 20° as the width of the upwelling branch of the Hadley cell circulation is in agreement with the observations of Kuhn et al. (to be published) and Kuhlbrodt (1931) as shown in figures 2 and 3. The resulting upward mass flux of air is $4.8 \times 10^{12} g\ s^{-1} (1.5 \times 10^{17} kg\ yr^{-1})$. On the basis of the recent findings of Kuhn et al., a mixing ratio in the rising part of the Hadley cell of 3.7 ppmm seems realistic. Thus, the moisture flux would be $1.7 \times 10^{7} g\ s^{-1} (5.5 \times 10^{14} g\ yr^{-1})$. It is envisioned here that the Hadley cell flux diffuses slowly through the tropopause without affecting its location, being determined by radiative parameters rather than by the dynamic conditions. For convective currents consequently, the tropopause still is a barrier.

2.2 Convective Storm Contribution to Stratospheric Moisture

Examples of tops of convective clouds at altitudes well above the tropopause are extensively documented. For example, Lee and McPherson (1971), in reporting their observations of thunderstorms over Oklahoma and Malaysia, noted that mean heights of tops in both areas during the study were 14.6 km (48,000 ft). Of 469 storms in Oklahoma with tops above 12.2 km (40,000 ft), 29 (6.2 percent) had tops in excess of 16.5 km

(54,000 ft), with 4 of these exceeding 18.3 km (60,000 ft). Of 62 Malaysian storms, 5 (8 percent) had tops above 16.5 km, but none exceeded 17.7 km (58,000 ft).

A tropopause height of 12.2 km over Oklahoma is common; whereas in tropical regions, such as Malaysia, the tropopause is usually at about 15.2 km (50,000 ft). Therefore, one can assume that every Oklahoma storm which creates cloud tops above 12.2 km has at least entered the transition zone, with most having penetrated into the stratosphere. According to Lee and McPherson (1971), about 20 percent of tropical thunderstorms rise into the stratosphere. From analysis of astronaut flight data, W. Shenk (personal communication) found the tops of Cb in a squall line in Oklahoma to be 21.9 km (72,000 ft). This water would be truly injected into the stratosphere above the altitude of the tropical tropopause. In inspecting figure 1, we call attention to the high mixing ratio found during the summer of 1965. This value was discarded by the author as instrument error. We must add the reminder, however, that on Palm Sunday in 1965, an extensive outbreak of highly intense convective activity occurred over Minnesota, Wisconsin, Illinois, Indiana, and Michigan that produced 34 destructive tornadoes. The storms which spawned these tornadoes probably reached very high levels and the possibility should not be discarded that they caused the high water-vapor mixing ratio.

That a cloud mass has entered the lower stratosphere does not mean, however, that the water contained therein is available for distribution within the stratosphere. It is entirely possible that most of the water merely sinks back into the troposphere when the storm decays. The mechanism by which the storms penetrate the tropopause is not entirely clear either; we hestitate to subscribe entirely to the parcel buoyancy theory, believing that changes in the stratospheric temperature structure due to strong radiation cooling effects of the spreading anvil play an important role in paving the way for subsequently rising storm turrets to reach the next higher level. The storm might be said to create its own tropopause when the thrust of the rising turrets pushes past the existing tropopause. Adiabatic expansion will create a region of very cold air that will subside to its equilibrium level when the storm updraft is removed. As an example, a typical Plains States tropopause is about 12.2 km, with a temperature of about 213°K. An air parcel carried to 15.2 km along the moist adiabat, without mixing and radiant energy exchange, would achieve a temperature of about 186°K. This agrees with an analysis by Roach (1967). Illustrative of this point is the temperature-pressure diagram for Topeka, Kans., on May 12, 1970, during a time when there was large-scale convective activity in the vicinity (fig. 5). Figure 6 (from Roach) is a structural visualization of the resultant cold dome over the active cell of a thunderstorm.

In such a situation as that just described, the uplifted cloud parcel could even act as a sink for stratospheric moisture. The density of water vapor at saturation over ice at 186°K is about 1.9×10^{-10}g cm^{-3}, whereas the water vapor density at 15.2 km, assuming a 3-ppmm mixing ratio, would be about 6.1×10^{-10} g cm^{-3}. This, obviously, is an extreme case in which there is no mixing. In reality, a wind shear almost always exists, and cloud parcels will mix with the ambient atmosphere which is much warmer.

In the following discussion, we estimate the vapor input into the stratosphere from the large thunderstorm of May 12, 1970, in northeast Kansas that was analyzed by Fujita (1972) (fig. 7). This storm developed during a period of about 2 hr. At 1845 local time, the anvil covered an area of about 60,000 km^2. Up to this time, several tornadoes had formed, three of which touched ground at least briefly. The tropopause was indicated at 11.9 km and had a temperature of 212°K. The greatest areal extent of the anvil occurred at 12.5 km, that height being 0.6 km above the tropopause. More than one-half the area of the cloud top was above 13.1 km, and several turrets rose from the anvil to altitudes as great as 15.8 km.

At the average altitude of the cloud top, ambient temperature was 213°K and pressure was 160 mb. If the cloud had risen above the tropopause along its moist adiabat, its temperature would have been 199°K. One can assume that mixing between cloud and ambient air produced a temperature at the interface of about 206°K and that this interface was about 100 m thick. The environmental air which experienced such turbulent

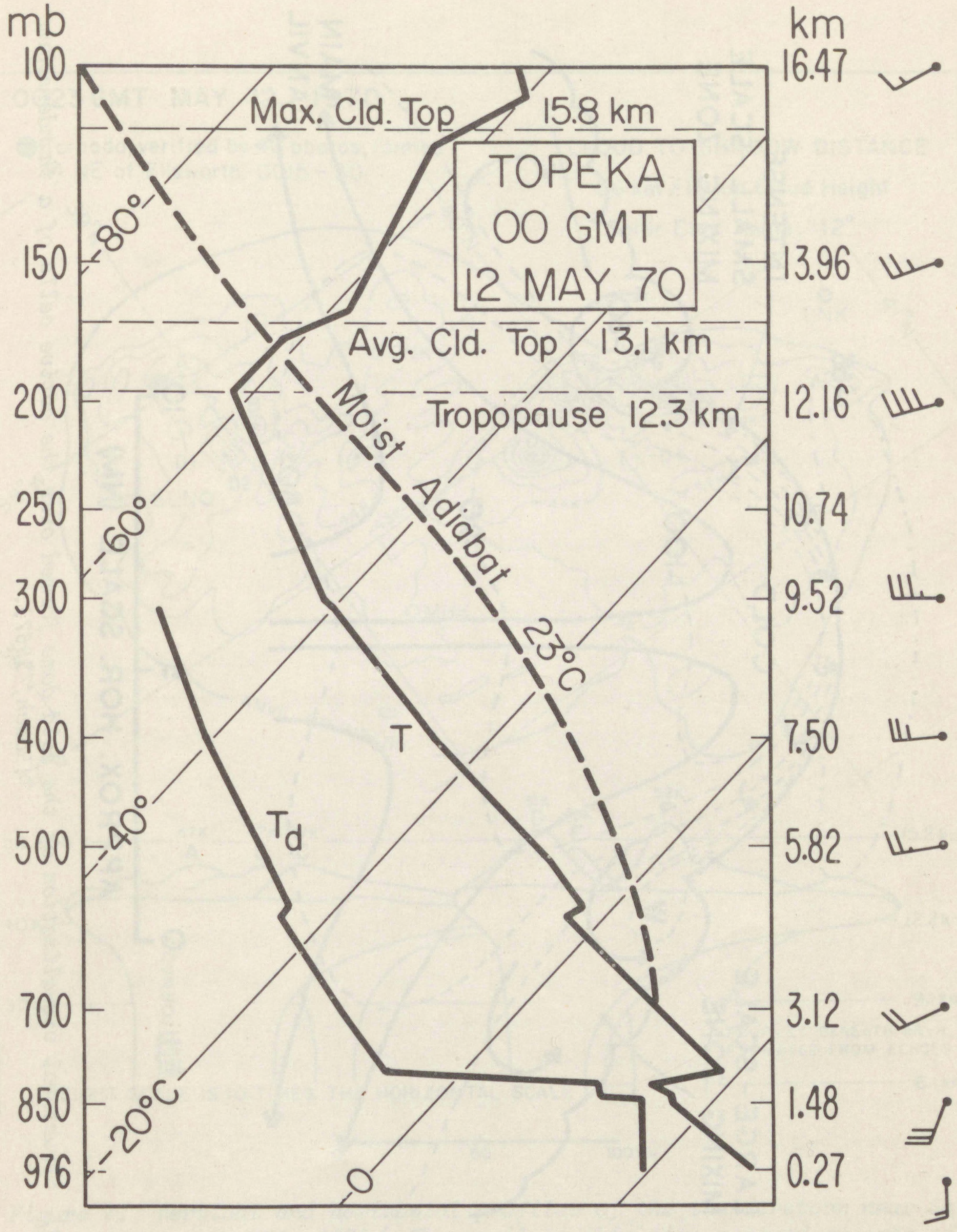

Figure 5. Temperature-pressure diagram for Topeka, Kans., during the existence of large-scale convective activity in the vicinity.

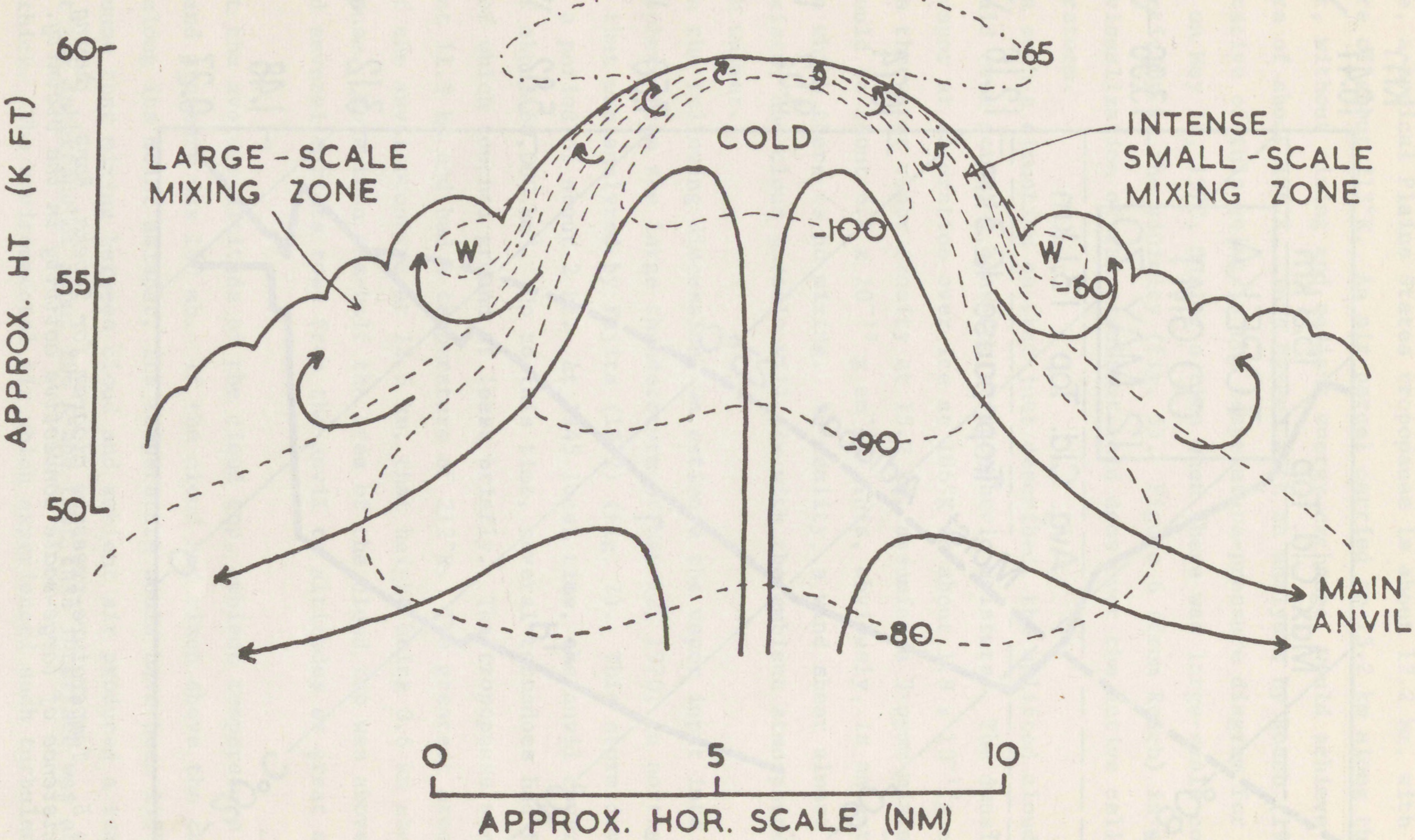

Figure 6. Structural visualization of the cold dome formed over the active cells of a thunderstorm (Roach, 1967).

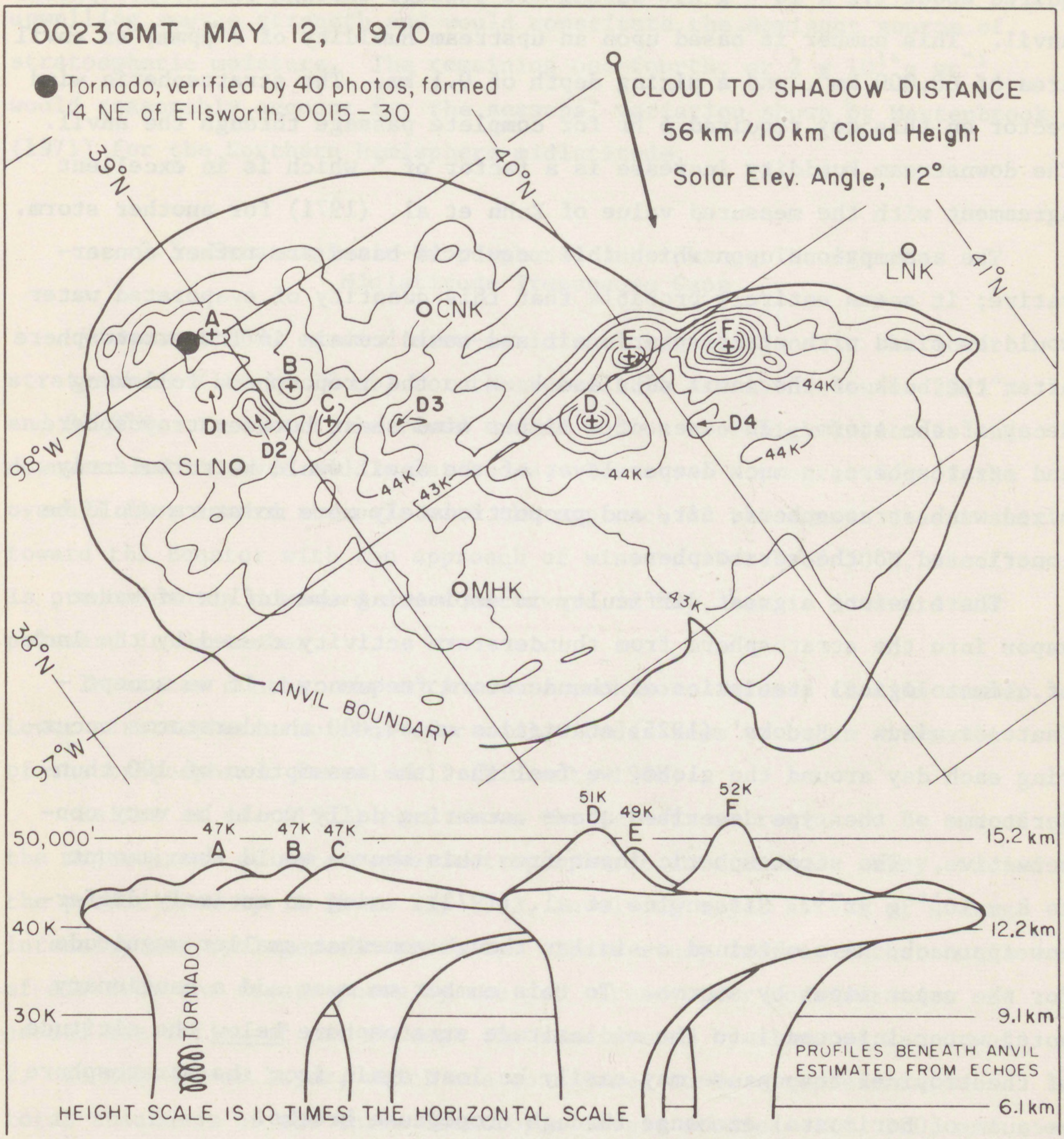

Figure 7. Vertical and horizontal profiles of the thunderstorm near Salina, Kans., May 12, 1970. The anvil at this time covered an area of about 60,000 km² (Fujita, 1972).

mixing within the interface assumed ice saturation and, therefore, acquired about 2.2×10^{10} g H_2O during its passage through the stratospheric anvil. This number is based upon an upstream humidity of 3 ppmm, an anvil area of 60,000 km^2, and a mixing depth of 0.1 km. The stratospheric wind vector on this day required 5 hr for complete passage through the anvil. The downstream humidity increase is a factor of 5 which is in excellent agreement with the measured value of Kuhn et al. (1971) for another storm.

The assumptions upon which this result is based are rather conservative; it seems entirely probable that this quantity of evaporated water would be mixed with stratospheric air and would remain in the stratosphere after the bulk of the anvil subsided back to the troposphere following decay of the storm. In cases of a higher wind shear between troposphere and stratosphere, a much deeper layer of the anvil would be turbulently mixed with stratospheric air, and proportionately more moisture would be contributed to the stratosphere.

There exists a great difficulty in estimating the influx of water vapor into the stratosphere from thunderstorm activity caused by the lack of climatological statistics of thunderstorm frequency. If we accept — faute de mieux — Brooks' (1925) statistics of 44,000 thunderstorms occurring each day around the globe, we feel that the assumption of 100 thunderstorms of the type described above occurring daily would be very conservative. The stratospheric input from this source would then amount to 8×10^{14} g yr^{-1}. Sissenwine et al. (1972), using an entirely different approach, have obtained a similar though somewhat smaller magnitude for the vapor input by storms. To this number we must add a cautionary note; water injected into the midlatitude stratosphere <u>below</u> the altitude of the tropical tropopause may easily be lost again from the stratosphere because of horizontal exchange through tropopause breaks.

An additional assumption is appropriate regarding the effective injection of moisture into the stratosphere because of thunderstorms. It should be recognized that most of the 4.4×10^4 daily thunderstorms occur over the continents and the large islands of the Tropics. We will assume, therefore, that three-fourths of the moisture injection from thunderstorms will occur in tropical regions and would be an addition to the Hadley circulation flux. The sum of the Hadley flux of 5.5×10^{14}g yr^{-1} and thunderstorm injection of 6×10^{14}g yr^{-1} would then represent the tropic-upwelling source strength and would constitute the dominant source of stratospheric moisture. The remaining one-fourth, or 2×10^{14}g yr^{-1}, would reasonably account for the seasonal variation shown by Mastenbrook (1971) for the Northern Hemisphere midlatitude.

2.3 Stratospheric-Tropospheric Interchange Through the Midlatitude Tropopause Gaps

Vincent (1968) studied mean-meridional circulations in the lower stratosphere (100 to 10 mb) for the years 1964 and 1965 between 22.5°N and 82.5°N. In winter, he found upward motion over high latitudes and downward motion over middle latitudes. There was a progression of the overall flow toward the pole with the approach of the summer season and toward the Equator with the approach of winter. North of 50°N, the flow is predominantly downward and equatorward in May, but is downward and poleward in September.

In winter months, the motions are much more intense than in summer. Lower stratospheric motions over midlatitudes seem to be an extension of the tropospheric Ferrel cell (Vincent, 1968).

Reiter et al. (1967) have shown the jet stream region to be one of the major areas of stratospheric-tropospheric interchange. They studied the period from April 18 to 21, 1963, involving a jet stream of average intensity and cyclogenetic activity. Their study indicated that outflow of air from the stratosphere by the jet stream under consideration exceeded the inflow of air through the same system by almost a factor of 2. In the 210-mb layer, which was considered to be a representative total thickness of the three isentropic levels studied—310, 320, and 330°K—the net outflow of air from the stratosphere was estimated to be 6.0×10^{16} g hr^{-1}.

The gross inflow of air, therefore, is inferred to be of similar magnitude. If we accept these figures to be realistic, that is, there is typically along a jet stream system a gross outflow of tropospheric air of 6×10^{16} g hr^{-1} and a gross outflow of stratospheric air of 12×10^{16} g hr^{-1}, then it becomes apparent that the net transfer of water vapor is likely to be upward. The reason is simply that the water vapor mixing ratio in upper tropospheric air is usually much more than double that in lower stratospheric air. If we assume a humidity of 10 ppmm for the outflow from the lower stratosphere and 50 ppmm for the outflow from the upper troposphere, the net transfer of water vapor to the stratosphere would be 18×10^{11} g hr^{-1}. The value of 50 ppmm for the upper troposphere amounts to 25-percent relative humidity at a temperature of -50°C. These assumed mixing ratio values are conservative, as is shown by the "excess frequency" of permanent vapor trails behind jet aircraft in the anticyclonic inflow side (Endlich and McLean, 1957). This indicates that ice saturation frequently occurs. At the temperature given, this is equivalent to 65-percent relative humidity.

Such an infusion of water vapor into the stratosphere during the winter period of maximum jet stream activity has not been reflected in the stratospheric mixing ratios as reported by Mastenbrook (1974) or others, that show maxima in late summer. This probably means that the mixing processes which transport water into the lower stratosphere are counteracted by other processes which return air to the troposphere poleward from the point of jet-stream intrusion, that is, mean meridional flow and subsiding air motion in high latitudes. These will be discussed later.

2.4 Methane Oxidation Contribution to Stratospheric Moisture

Methane is produced at the earth's surface at a rate of 2 to 3×10^{11} molecules cm^{-2} s^{-1} (Robinson and Robbins, 1968; Wofsy et al., 1972) and is present in the troposphere at about 1.0 ppmm (Bainbridge and Heidt, 1966; Kyle et al., 1969; Ehhalt, 1967; Robinson and Robbins, 1968). The mixing ratio above the tropopause has been found to decrease with altitude, reaching values of 0.5 ppmm at 30 km and of 0.15 ppmm in the vicinity of the stratopause (44 to 62 km) according to Ehhalt and Heidt (1973) and Ehhalt et al. (1972), respectively. Ackerman and Muller (1973) obtained mixing ratios in general agreement, namely, 1.3 ppmm at 15.9 and 24.6 km, 0.7 at 29.6 km, 0.6 at 31.9 km, and 0.4 at 33.6 km.

Destruction of methane in the stratosphere occurs by oxidation, beginning primarily with an attack by the hydroxyl radical to produce the methyl radical and a molecule of water,

$$CH_4 + OH \rightarrow CH_3 + H_2O \ .$$

In the upper part of the stratosphere, the reaction with excited oxygen atoms becomes more important,

$$O(^1D) + CH_4 \rightarrow OH + CH_3 \ .$$

The ultimate result of completion of the reaction chain begun with these reactions is the formation of one molecule of carbon dioxide and two molecules of water for the destruction of a molecule of methane. In consequence, the stratospheric water-vapor mixing ratio increases with altitude. The oxidation of methane occurs in several steps, with important atmospheric trace gases such as formaldehyde, carbon monoxide, and methyl hydroperoxide occurring as intermediates (Nicolet, 1972; Nicolet and Peetermans, 1973; Crutzen, 1973; Warneck et al., 1973).

Although large-scale organized vertical motions may play a part in the upward movement of methane within the stratosphere, their effect is likely to be small because of the very rapid and efficient lateral mixing (Warneck et al., 1973).

Several mathematical models have been constructed to aid in specifying the sources, reactions, and distribution of stratospheric trace gases. Wofsy et al. (1972) assumed a net upward flux of 4 x 10^9 methane molecules cm^{-2} s^{-1} at the tropopause and 2 x 10^9 molecules cm^{-2} s^{-1} at 25 km; they then accepted an eddy diffusion coefficient of 10^4 cm^2 s^{-1} between 25 and 40 km to meet the observed mixing ratio at 44 to 62 km of 0.15 ppmm (Ehhalt et al., 1972). Nicolet (1972) and Nicolet and Peetermans (1973) stated that the upward current of methane molecules across the tropopause could not be less than 5 x 10^9 molecules cm^{-2} s^{-1} which corresponds to a stratospheric production of not less than 10^{10} water molecules cm^{-2} s^{-1}. For this stated minimum flux, the model of Nicolet and Peetermans requires their maximum eddy diffusion coefficient which varies from ~1.5 x 10^5 cm^2 s^{-1} at 10-km altitude to ~1.5 x 10^4 cm^2 s^{-1} at 35 km and 4 x 10^4 cm^2 s^{-1}

at 50 km, as well as a ratio of hydroperoxyl (HO_2) to hydroxyl radicals of 9 and a solar zenith angle of 60° to approach the mixing ratio found by Ehhalt et al. from 44 to 62 km. These model conditions satisfy the mixing ratio observations of Ackerman and Muller (1973) that were obtained at altitudes up to 33.6 km. However, the minimum tropopause flux used in this example would correspond to an eddy diffusion coefficient across the tropopause of 2×10^3 cm^2 s^{-1} (Nicolet) instead of the 1.5×10^5 cm^2 s^{-1} used just above the tropopause. These are, of course, limiting conditions; a larger flux coupled with a smaller eddy diffusion coefficient would probably come closer to reality. Ehhalt and Heidt (1973) derived a flux of 2.5 to 5×10^{10} molecules cm^{-2} s^{-1}. Crutzen (1973) used a vertical eddy diffusion coefficient of 10^4 cm^2 s^{-1} to operate between 10 and 50 km. His lower boundary mixing ratio was 1.0 ppmm at 10 km, with an upper boundary of 0 at 90 km. The upward flux condition of 6×10^9 methane molecules cm^{-2} s^{-1} at 10 km was coupled with a rather fast rate for $OH + O_3 \rightarrow HO_2 + O_2$ ($k = 2.5 \times 10^{-11} \exp(1500/T)$). This model reasonably approximates the observed concentrations of methane of Ackerman and Muller and of Ehhalt and Heidt in the 20- to 34-km range, but predicts a lower concentration at the 40- to 50-km range than was found by Ehhalt et al. For the above reaction, a rate of $k < 10^{-16}$ is also listed by Crutzen. If employed, this rate would reduce the reaction to insignificance and allow one-half an order-of-magnitude increase in hydroxyl in the middle stratosphere. The increased hydroxyl concentration would result in more rapid destruction of methane, and the model would then require a greater flux of methane at the lower boundary or a larger eddy diffusion coefficient, or both.

Eddy diffusion coefficients on the order of 10^4 cm^2 s^{-1} are most in favor, and it seems reasonable that the flux of methane across the tropopause is about 1.5×10^{10} molecules cm^{-2} s^{-1}. This flux would ultimately result in the addition of 3×10^{10} water molecules cm^{-2} s^{-1} (1.4×10^{14} g yr^{-1}) to the stratosphere.

2.5 Volcanic Activity as a Source of Water Vapor

It has been long recognized that water is a major constituent of volcanic gases, and the force required to produce an explosive eruption is thought to derive from a release of pressure sufficient to permit vaporization of the water in the magma (Bullard, 1962, pp. 37-43). Day and Shepard (Bullard, loc. cit.) in 1912 determined that steam averaged 68.2 percent by volume in the gases of Kilauea Crater, which at that time had a permanent lake of molten lava. More recently, Heald et al. (1963) used more modern techniques to analyze the gases from Kilauea during and following the eruptions of 1959 and 1960. They found water and carbon dioxide to be the dominant gases, with most samples containing water at more than 90 mole-percent.

The volcanic cycle beginning in 1963 has seen eight eruptions in which the eruption cloud entered the stratosphere. In March 1963, Agung volcano on the island of Bali erupted in the single most significant explosive event since Bezymianny in 1956. A month later, Trident in Alaska erupted, spewing gas and ash clouds to 15-km altitude; and in November 1963, Surtsey began to form in the sea off Iceland. Surtsey exploded with the greatest violence when the crater was open to the sea, but the eruption cloud rose to the greatest altitude after the crater rim blocked entry of the water. Surtsey's eruption cloud apparently rose to about 50,000 ft (Cronin, 1971). Other eruptions which ejected gas and ash into the stratosphere were: Taal volcano in the Philippines in September 1965, Redoubt in Alaska in January and February 1966, Fernandina in the Galapagos Islands of Ecuador in May 1968, Hekla in Iceland in May 1970, and Haemaey in Iceland in April 1973.

Although all volcanoes emit considerable amounts of water, three of these eight eruptions--Surtsey, Taal, and Fernandina--were phreatic or submarine explosions and may have injected significant quantities of water vapor into the atmosphere. The first 3 months of Surtsey's eruption, when sea water had access to the tephra walls, may very well have been a source of stratospheric water for the northern stratosphere. It is interesting that Mastenbrook's (1971) data show no special H_2O increase in 1964, except for a high value during mid-1964 several months after the Surtsey eruption. Of course, this high value is considered by the author to be a measuring error.

Wilson et al. (1966) estimated the volume of gas ejected during the climactic eruption of Redoubt to be 9 km^3. If, to conjecture an extreme situation, one assumes that Surtsey, Taal, or Fernandina had ejected 10 km^3 of water vapor, beginning at a density of about 1 kg m^{-3}, to stratospheric altitudes, the resulting mass of water in the stratosphere—about 10^{13} g—would probably have been notable before dispersion. However, following dispersion, but assuming no loss, the water density from the volcanic injection would be only 1 percent of the normal stratospheric water vapor. From this, it seems very unlikely that volcanic eruptions during the recent period of activity have in any significant way affected stratospheric water vapor concentrations.

3. REMOVAL OF STRATOSPHERIC MOISTURE

Above approximately 70 km, water vapor is destroyed through photochemical processes, largely through photodissociation by solar ultraviolet of wavelength <2340Å. But the equilibrium between water vapor and its products of dissociation is not altered, except in terms of geologic time, by the very low loss rate of protons to space (10^8 cm^{-2} s^{-1}).

According to Hesstvedt (1968), the actual water vapor density in the vicinity of the mesopause, as inferred from the observation of noctilucent clouds, is approximately two orders of magnitude higher than the water vapor density at photochemical equilibrium. Thus, for the summer mesopause, mechanisms for upward transport of water vapor are necessary; mechanisms for downward transport are required for the winter mesosphere.

Because water is not consumed above the stratosphere and because the stratosphere is actually a source for water vapor through the oxidation of methane, significant mechanisms for removal from the stratosphere must lie in the large-scale circulation of the atmosphere. Thus, removal of water vapor occurs with airmass removal in the downward-moving branch of the Hadley cell circulation. The early discharge of Tungsten 185 at 20°S and 30°N, several weeks after its infusion into the tropical tropopause (Newell, 1962), is evidence of the effectiveness of this mechanism.

The katabatic winds at the fringes of the Greenland and Antarctic ice domes may be evidence of additional substantial sinks for stratospheric moisture. Particularly over the Antarctic plateau, the tropopause is poorly defined or nonexistant and can be said, with some justification, to intersect the surface. This circumstance, coupled with the persistent sinking motions throughout the stratosphere over this region, as postulated by Rieter et al. (1967) and by Reiter (1971), would provide a means by which the stratosphere could supply much of the moisture and air for surface accumulation and outflow across the continental margin. According to Loewe (1972), the quantity of snow transported across parts of the Antarctic coastline is considerable, reaching 7×10^{15} g yr^{-1} for 100 km of coastline in the vicinity of Cape Denison and Port Martin (67°'S, 142°41'E, and 66°49'S, 141°24'E, respectively). Along this part of the coastline, the loss by wind action exceeds the loss by iceberg formation and by melting. The yearly mean windspeed, 18 m s^{-1} (gale force), along this part of the coastline is not equalled along other sectors of the Antarctic coast, but most other sectors of the 15,000-km coast experience gale force katabatic winds 5 to 10 percent of the time (Lamb, 1957).

The dominantly zonal flow of the winter Antarctic stratosphere and general subsidence over the Antarctic continent would feed stratospheric components into the outflowing surface winds. In addition, an important sink function that is specific for water vapor is attributed to the Antarctic stratosphere because of the extremely low temperatures during the winter months (Ellsaesser, 1974). During this period of time, the saturation mixing ratio is often less than the mixing ratio of inflowing stratospheric air, and the excess water vapor is removed by formation and settling of ice particles. Stanford (1973) analyzed radiosonde data for 50 mb at the South Pole for 11 years; he found that the mean mixing ratio at saturation with respect to ice was about 1 ppmm during August. The values for June and July were about 2.8 and 1.6 ppmm, respectively. Mean mixing ratios at saturation with respect to ice at 100 and 150 mb were also less than 3 ppmm during a 2- to 3-month winter period. The stratospheric clouds expected with such temperatures were observed by members of the Norwegian-British-Swedish 1949-52 expedition to Antarctica (Liljequist, 1956). The "cloud-veil" was seen day after day from June to the beginning of October.

A similar condition exists in the circulation over Greenland (Weickmann, 1961). Figure 8 shows the wind direction and velocity at several surface stations, including the predominant outflows from the icecaps. Figures 9 and 10 give wind observations from the surface to 10 km (tropopause level at about 8 km) at Site II (west coast) and Danmarkshavn (east coast), respectively. On the west coast of the ice cap, upper level winds are predominantly WNW to NNE (fig. 9), while on the east slope, the upper level winds are from the SW. This wind direction indicates outflow at low levels from the ice cap and inflow aloft toward the ridge of the ice cap. Continuity calls for subsidence over the ice cap. Unfortunately, the amount of wind data is too limited to permit a sufficient analysis of air parcel trajectories to compute the loss of moisture from the polar stratosphere.

North polar stratospheric temperatures are higher than Antarctic stratospheric temperatures. According to a review of observations at Jan Mayen Island (70°56'N, 08°40'W), the 30-mb temperature is -80°C or lower on 6.4 percent of the days in December and on 18.0 percent of the days in January (Stanford, 1974). Therefore, assuming a similar water-vapor mixing ratio for stratospheric air approaching Greenland as for Antarctica, the formation of ice clouds will occur much less often over Greenland, and the specific sink for water vapor through settling of ice crystals will be correspondingly less important.

The apparent existence of an average annual north-to-south gradient in stratospheric water vapor, seen in an extreme form by McKinnon and Morewood (1970), is further indication of the important sink function of the Antarctic stratosphere (Ellsaesser, 1974). It is strongly suggested that ice crystal formation over the Antarctic and Greenland ice domes comprises the only important sink that is specific for stratospheric moisture.

Figure 8. Surface wind direction and velocity at several Greenland stations (Weickmann, 1961).

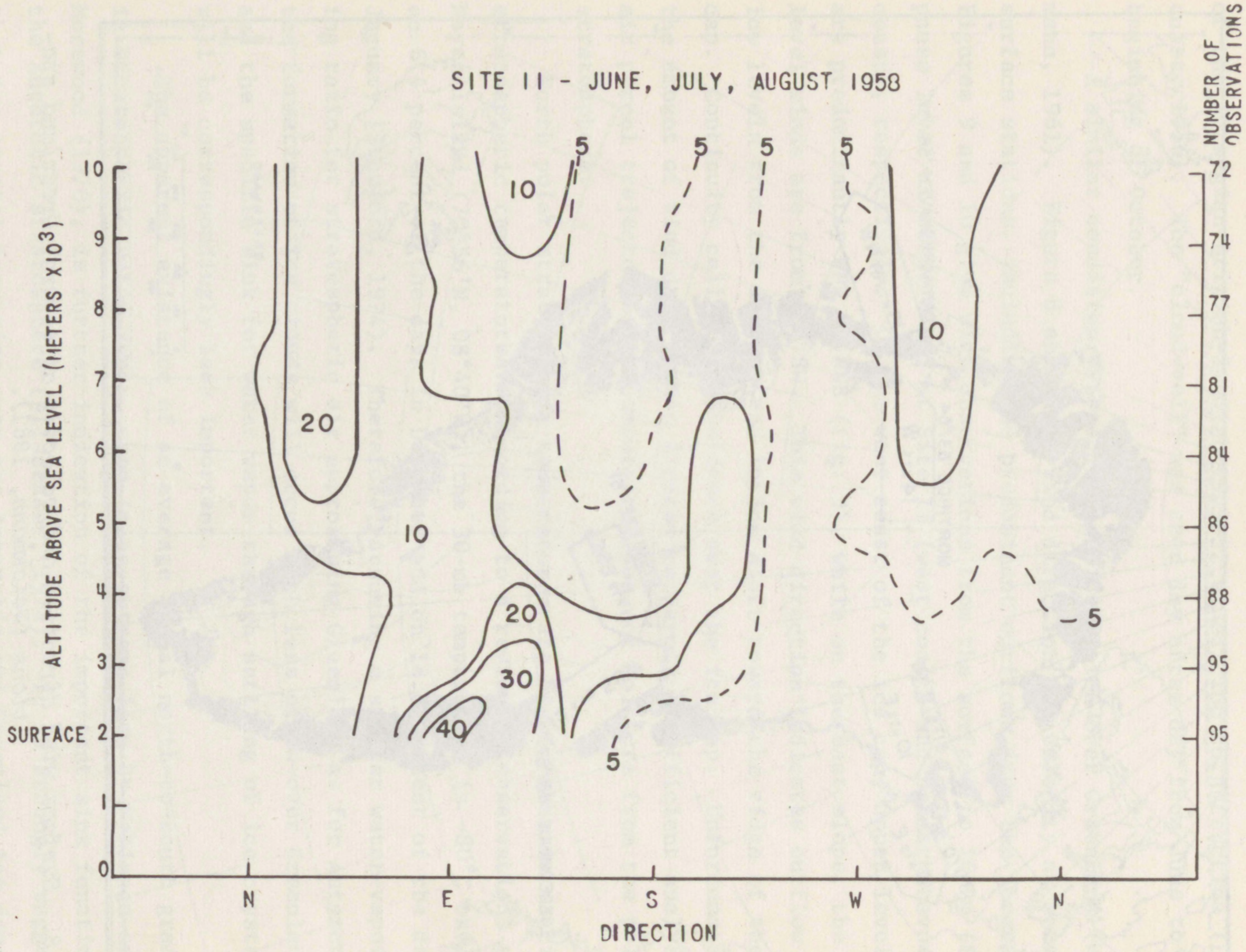

Figure 9. *Wind observations from the surface to 10-km altitude at Site II (west coast), Greenland. (H. K. Weickmann, unpublished).*

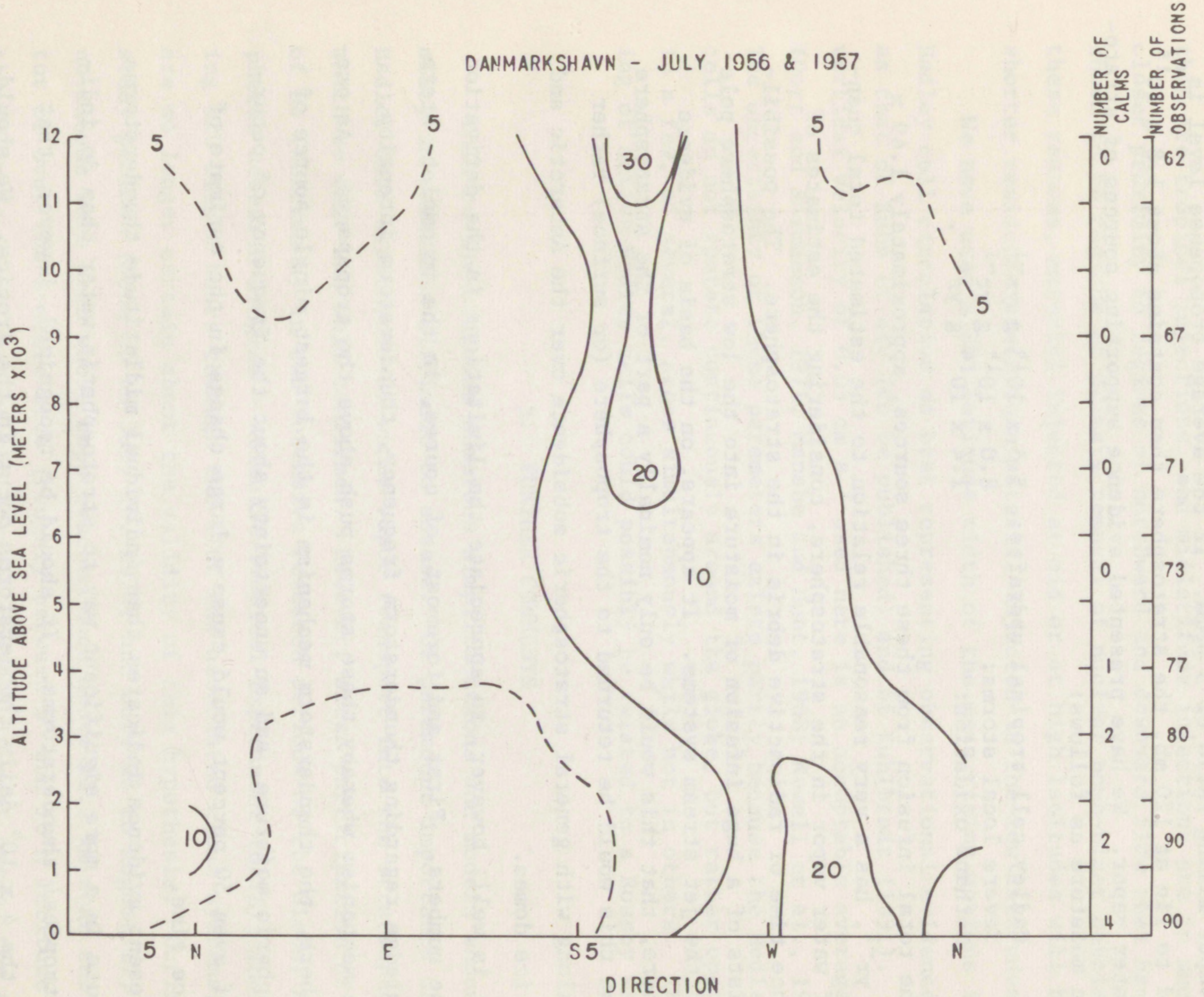

Figure 10. *Wind observations from the surface to 12-km altitude at Danmarkshavn (east coast), Greenland. (H. K. Weickmann, unpublished).*

4. CONCLUSION

Present evidence supports the acceptance of an average stratospheric water-vapor mixing ratio of 3 ppmm. If the average tropopause level is assumed to lie at 120 mb, the stratosphere then contains about 1.8×10^{15} g of water vapor. We have presented evidence supporting sources of stratospheric moisture as follows:

Hadley cell tropical updrafts:	5.5×10^{14} g yr^{-1}
Severe local storms:	8.0×10^{14} g yr^{-1}
Methane oxidation:	1.4×10^{14} g yr^{-1} .

The total infusion from these three sources, approximately 1.49×10^{15} g yr^{-1}, has a very reasonable relation to the estimated total quantity of water vapor in the stratosphere, considering the estimates of residence time of radioactive debris in the stratosphere. The possibility exists of a huge infusion of moisture into the low stratosphere poleward of the jet stream systems. It appears, on the basis of evidence cited here, that this would be only nominally a part of the stratosphere and that this would be returned to the troposphere (or surface) rather soon, along with general stratospheric subsidence over the Antarctic and Arctic ice domes.

It is well, however, to appreciate the limitations in the derivation of these numbers. First and foremost, of course, is the incomplete state of knowledge regarding thunderstorm frequency, thunderstorm intensity, and the mechanism whereby these storms push above the tropopause. As presented here, the thunderstorm mechanism is the largest single source of stratospheric moisture, and an uncertainty about the frequency of occurrence of even 50 percent would cause a large change in the estimate of residence time.

Present evidence indicates that individual midlatitude thunderstorms contribute in a more significant way to stratospheric water than do individual tropical thunderstorms. It should be recognized, however, that most of the 4×10^4 daily thunderstorms occur in the Tropics. We should, therefore, add a large fraction, perhaps three-fourths, of the storm injection to the "dry" Hadley circulation flux and treat the sum as the total tropical-upwelling source strength; this sum would then constitute the dominant source. In addition, injections in midlatitudes are much more likely to occur in areas of general subsidence of stratospheric air than are tropical injections, and midlatitude injections are in much closer proximity to regions of northward and downward flow that have been shown to be regions of primary removal of nuclear bomb-blast debris. For these reasons, material injected at mid or at high latitudes will have shorter residence times than material injected in the low latitudes.

We have used a 20° latitude width of the mean rising motions in the Hadley cell circulation as best representing observational evidence such as that of Kuhn et al. (to be published) and of Kuhlbrodt (1931). The vertical velocity of 0.03 cm s^{-1} used here is an acceptable average value (Oort and Rasmusson, 1971; Manabe and Hunt, 1968; Newell et al., 1969). The uncertainty of these parameters exists partly because the Hadley cells do not operate continuously around the globe, but rather operate in a few, or several, areas simultaneously while not in others. The spacing of active Hadley cells could possibly be related to a Rossby wave.

5. FURTHER COMMENTS

This review contains numerous simplifications and models whose reality is sometimes difficult to assess. Of paramount importance is the Hadley cell hypothesis of water vapor transport into the stratosphere which we felt was well confirmed. However, after participation of one of us (Weickmann) in two intensive research periods of the Global Atmospheric Research Program — Atlantic Tropical Experiment (GATE), including 120 flight hours through the ITCZ and associated cloud systems, we are no longer certain about the validity of this hypothesis. During the study periods (June 26 to July 16, 1974, and July 28 to August 17, 1974), no clouds were seen whose tops reached much above 12.2 km near the Equator or over the B-scale area (roughly between 5° and 10°N latitude over the Atlantic Ocean west of the African coast); in most cases, the air above the clouds appeared to be very dry as indicated by the frequent lack of aircraft condensation trails. The altitude of the tropopause was, for the most part, about 15.2 km. The concept of the Hadley cell which we have utilized involves a more or less continuous band of rising motion around the world (Newell et al., 1969). However, if the lack of high-altitude clouds over this part of the tropical Atlantic Ocean is typical of significant parts of the tropical open ocean, then the role of high-reaching convective storms over the tropical land areas must be much more significant than has heretofore been recognized. We hope that high-resolution satellite pictures may in the future enable us to assess more accurately the number and size of high-reaching convective storms and to arrive at an answer to the important question of what is the role of the Hadley cell for the vertical transport of water vapor.

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