A Thin Stable Layer of Anomalous Ozone and Dust Content

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ABSTRACT

Coincident observations of a layer of volcanic material and a sharp minimum in the vertical distribution of ozone over Boulder, Colo. (40N), are presented and discussed. The ozone minimum was observed at an altitude of 20–21 km for a month during the spring of 1964.

Quasi-horizontal advection of a thin stable layer of tropical air into which volcanic debris was injected when Mt. Agung (8S) erupted on 17 March 1963 is thought to be responsible. The layer is characterized by a "quasi-vertical" eddy diffusion coefficient less than or equal to 2.5×10^2 cm^2 sec^{-1}. Significant destruction of ozone by the volcanic debris is not indicated.

1. Introduction

The development and improvement of ozone sounding techniques in recent years (Hering and Dütsch, 1965) makes it possible to study the distribution of ozone with much greater vertical resolution than in the past. This has provided further evidence of the highly stratified nature of the lower and middle stratosphere, confirming that of other tracers such as potential temperature (Danielsen, 1959), radioactivity (Anderson, 1965) and dust (Bigg, 1964; Newkirk and Eddy, 1964).

In this paper coincident observations of a thin layer of volcanic debris and a sharp minimum in the vertical distribution of ozone are presented. It is inferred from these and various supplementary observations that a thin layer of tropical air was advected nearly horizontally to middle latitudes without undergoing significant mixing with the surrounding air.

2. Ozone observations

An examination of routine ozone soundings made at Boulder, Colo., in March and April of 1964 with Brewer ozonesondes of the bubbler type (Brewer and Milford, 1960), manufactured by Mast Development Company, revealed an unusually sharp and persistant minimum in the ozone content at the 50-mb level (20.6 km altitude). This is illustrated by the five consecutive daily distributions shown in Fig. 1.

The dip persisted, as indicated in Fig. 2, from 9 March to 10 April, during which period 23 soundings were made. The mean ozone distribution around the 50-mb level is shown in Fig. 3.

A similar minimum in the ozone concentration was observed in soundings made at Fort Collins, Colo., 18 and 25 March, Albuquerque, N. Mex., 18 and 25 March and 8 April, and at Bedford, Mass., on 1 and 2 April (Hering and Borden, 1965). These latter soundings were part of the AFCRL ozone sounding program and were made with Regener chemiluminescent ozonesondes.

Comparisons between different sondes and comparison of ascent and descent data lead us to be confident of the detailed shape of the distributions obtained at Boulder, at least for features down to a thickness of a few hundred meters in the vertical.

3. Optical observations

Twilight balloon observations (Pittock, 1963) were made at Boulder during the evening of 30 March 1964. These consisted of the measurement, in arbitrary units, of the intensity of the narrow cone of rays from the sun which passed tangentially through the atmosphere at some minimum height $h_{\min}$, and which was reflected by a large meteorological balloon floating at an altitude of about 33 km. The intensities were measured using a filter photometer mounted on a telescope as described in the earlier paper.

As in the cases discussed by Harris (1964), increased attenuation in the tangential path prevented the derivation of an ozone distribution by the twilight balloon method at Boulder. However, measurements of intensity in the 0.44-μ band (Fig. 4) permitted derivation of the distribution of abnormally attenuating material.

In the absence of dust in the stratosphere one would expect the intensity to decrease approximately exponentially due to Rayleigh scattering by the increasing air mass in the tangential path (ignoring the negligible effect of ozone and other molecular absorption at 0.44 μ). Such was the case in the observations reported by Pittock (1963). It appears from these earlier Melbourne results that stratospheric aerosol sizes and number distributions (Junge *et al.*, 1961) are normally such as to have a negligible effect on the measurements at 0.44 μ.

Curves corresponding to Rayleigh scattering have been computed using air mass tables by Link and Neuzil (1965) and are shown in Fig. 4. The Boulder observations show a marked increase in attenuation, beyond

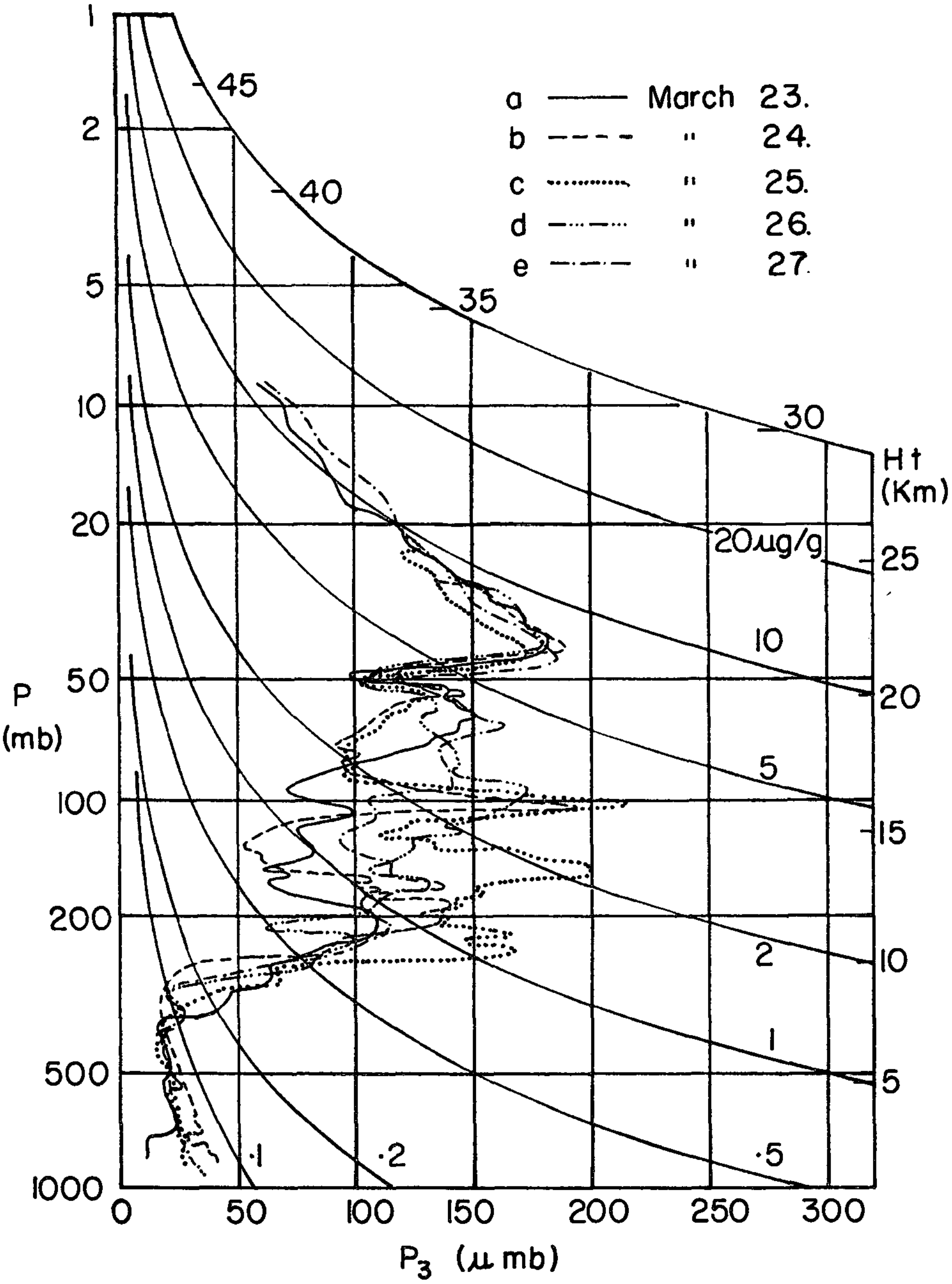

FIG. 1. Complete ozone soundings (ascents only) for the five consecutive daily flights from 23 through 27 March 1964 at Boulder, Colo. Lines of equal mass mixing ratio are indicated.

that expected due to Rayleigh scattering, for minimum heights of the tangential ray below about 21.3 km.

4. Nature and distribution of attenuating material

Besides its intrinsic interest, an assessment of the nature and distribution of the attenuating material should throw light on the coincident minimum in the ozone distribution observed over Boulder.

A series of observations of sunset, twilight and absorption effects (Meinel and Meinel 1963, 1964; Flowers and Viebrock, 1965; Volz, 1964, 1965) in the Northern Hemisphere strongly suggest that the abnormal attenuation reported above was due to material originating in the eruption of Mt. Agung on Bali (8° 25′S, 115° 30′E) on 17 March 1963.

Hence, as a first approximation, it seems reasonable to assume the same particle size distribution over Boulder as was found at the same time over Australia by Mossop (1964). Allowing for a coating of water-soluble material, we therefore assumed a mean particle diameter of 1 μ.

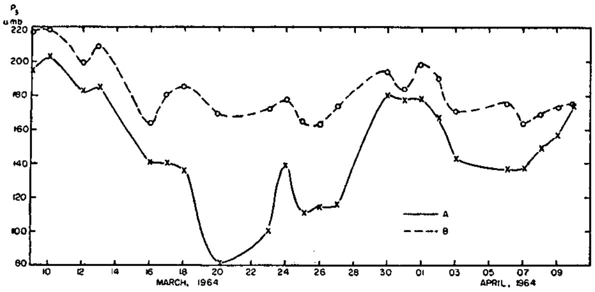

FIG. 2. Partial pressure of ozone at 50 mb at Boulder, Colo., during the period 9 March to 10 April 1964. Curve A represents the observed values while curve B represents the hypothetical ozone amount at the 50-mb level which would have been present if the minimum had not been present, using linear interpolation between the adjacent maxima.

On the basis of Mie scattering theory, the geometry of the situation (taking uniform layers 1 km thick) leads to the vertical distribution of attenuating particles shown in Fig. 5. This result is not very dependent on the assumed refractive index and is of rapidly decreasing accuracy below 19 km altitude.

As the path length through the dust layer is some hundreds of kilometers, Fig. 5 may be taken as representative of the attenuating layer over a large area.

The particle concentrations derived here are consistent with the effects reported by Flowers and Viebrock (1965) for Hawaii (20N) and Dyer and Hicks (1965) for Aspendale (38S), considering the latitudinal and time variations one might expect on the basis of studies of other tracers (Bolin, 1964).

The concentrations found over Boulder are sufficient to account for about one quarter of the total dust content over Hawaii. This suggests that in the narrow height range observed over Boulder the dust was transported with very little dilution from 20N.

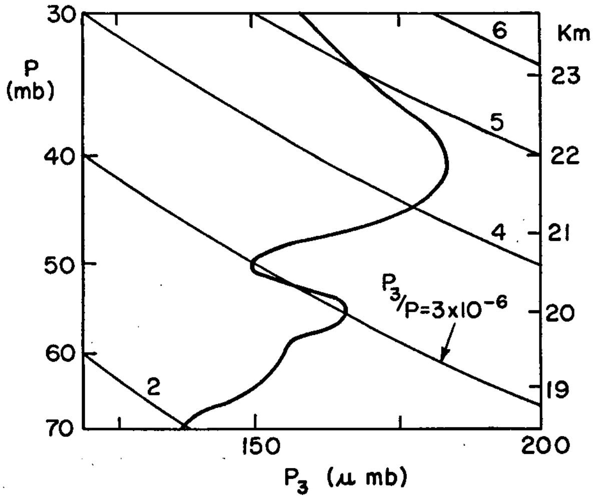

FIG. 3. Mean partial pressure of ozone vs. pressure, between 30 and 70 mb for all 23 consecutive soundings from 9 March through 10 April 1964, at Boulder. Lines of equal molecular mixing ration are indicated.

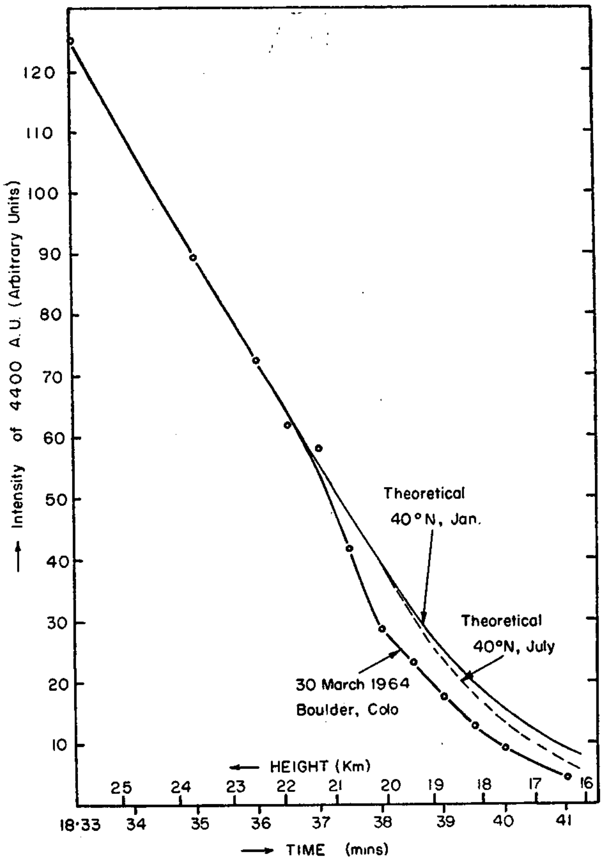

FIG. 4. Intensity of light from the balloon in the 0.44-μ band, plotted against local time and the minimum height of the tangential ray $h_{\min}$. The heavy line represents the observations made at Boulder on 30 March 1964, while the other two lines represent theoretical Rayleigh scattering curves for 40N, based on tables by Link and Neuzil (1965). All three curves are reduced to the same arbitrary units of intensity at $h_{\min}=25$ km.

5. Discussion

The ozone mixing ratio at levels around 20 km is nearly conservative, Dütsch (1956) finding a half-restoration time of about two years. It follows (Reed, 1950) that the pronounced ozone minimum reported above could not result solely from vertical mixing or subsidence. However, it could result from quasi-horizontal advection of a thin layer of tropical air to 40N.

Fig. 6 shows the mean of the "provisional" ozone distributions through March and April 1963 as determined at the Canal Zone station (9N) of the AFCRL ozone sounding network (Hering, 1964; Hering and Borden, 1964). It can be seen that tropical air below, at, or even a kilometer or so above, the 50-mb level has an ozone mixing ratio less than or equal to that observed at the 50-mb level over Boulder in March and April 1964.

The suggested origin of the attenuating material observed over Boulder, namely the eruption of Mt. Agung at 8S in March 1963, strongly supports the suggestion that air of tropical origin was advected as far as Boulder. The observations are not sufficient to describe the proc-

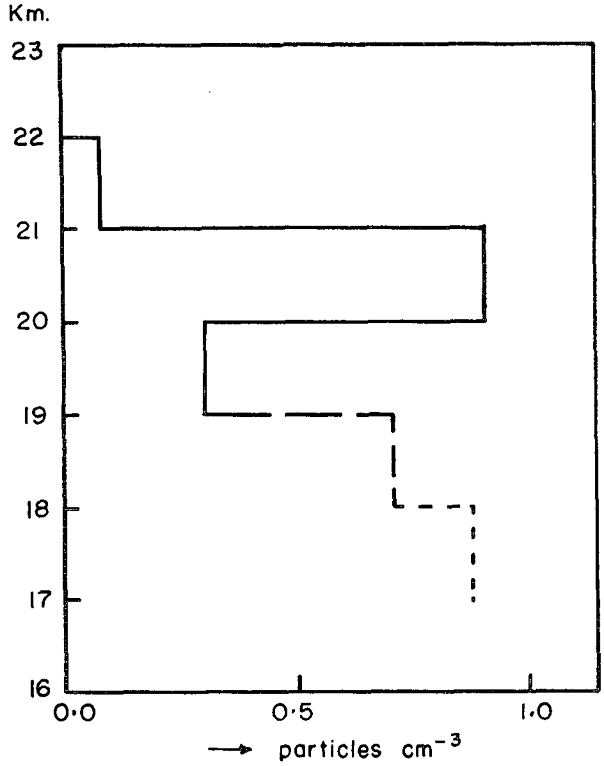

FIG. 5. Vertical distribution of 1-μ diameter particles derived from the observations of Fig. 4. The distribution is of rapidly decreasing accuracy below 19 km altitude.

ess in detail but they are indicative of several of its aspects.

Occasional sharp minima in the ozone content at the 50-mb level were observed over Boulder from December 1963 through February 1964. These may have been due to other intrusions of tropical air, but could have been due to the same lamina which was almost stationary over Boulder in March and April. During these latter months the wind at 50 mb went through its seasonal shift from westerly to easterly, the mean zonal component from 9 March to 10 April being 2.3 m sec^{-1} (easterly). This suggests that the ozone-depleted layer had a horizontal extent of the order of a few thousand kilometers. The observation of the layer at Albuquerque and Bedford, but not at other ozone network stations in the same latitude range, is consistent with this estimate.

The quasi-horizontal nature of the advection is supported, though not conclusively proved, by the fact that the ozone minimum at Albuquerque (35N) and Bedford (42N), and the upper boundary of the main dust layer at Tucson (32N) (Meinel and Meinel, 1963), were all between 20 and 21 km altitude. The lower heights included by Meinel and Meinel probably reflect the influence of horizon obscuration of the sun by distant cloud layers (personal communication, 1964).

Unless ozone was destroyed catalytically by the volcanic material, as suggested in an earlier note (Pittock, 1965), destruction cannot have played an important role in producing the observed ozone minimum since the observed amount of debris over Boulder could have caused at most less than 1 per cent of the ozone to be destroyed by oxidation-reduction processes. As destruction is not required to explain the observations, while advection is necessary, significant ozone destruction is not indicated.

The suggested quasi-horizontal advection of a thin layer of tropical air to 40N implies a remarkable lack of mixing of the layer in question with the air above and below it. Unfortunately, we are not able to say on the basis of the observations exactly how long this 0.5 km thick layer retained its identity, but we can fix a lower limit of about one month.

Simple diffusion theory requires, for a layer of thickness z to substantially retain its identity over a time t, a diffusion coefficient K_z given by

$$z^2 = 4K_z t.$$

The observations therefore require a value of K_z less than or of the order of 2.5×10^2 cm^2 sec^{-1}.

As most other investigators have estimated values of the vertical eddy diffusion coefficient in the middle stratosphere of about 5 or 10×10^3 cm^2 sec^{-1} (Newkirk and Eddy, 1964), the peculiar characteristics of the present estimate are of interest.

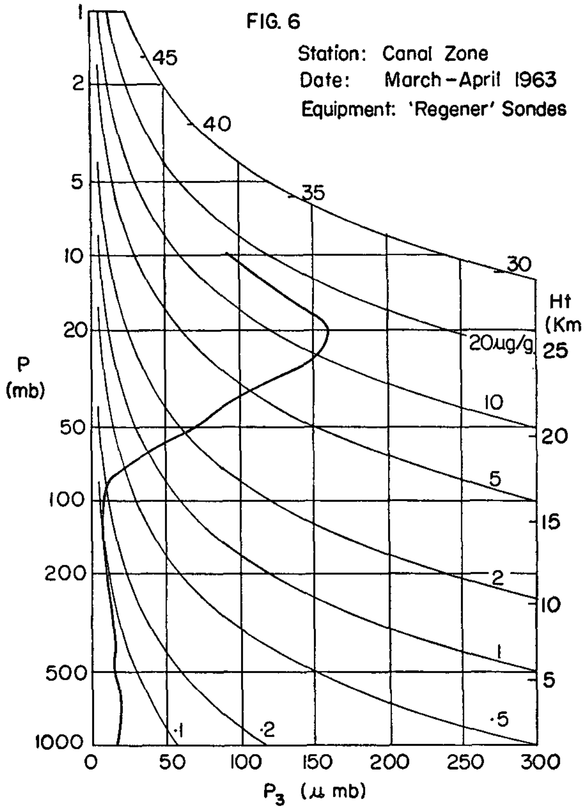

FIG. 6. Mean of seven provisional ozone distributions from the Canal Zone station of the USAF ozone sounding network during March and April 1963.

1) We are considering a lamina which stood out essentially because of the unusual length of time during which it retained its identity. We should, therefore, expect it to have a value of K_z lower than the mean value.

2) In so far as we may consider the lamina to have persisted for a period greater than one month, with a correspondingly smaller value of K_z, we in effect ignore any mean vertical motion of the lamina between the tropics and 40N. If we regard the suggested quasi-horizontal advection as being part of a large-scale quasi-horizontal eddy along a mixing path of slope α (Newell, 1963; Reed and German, 1965), it is clear that we are ignoring the contribution to the large-scale vertical eddy flux of the mean slope $\bar{\alpha}$ of the mixing path. The "vertical" eddy diffusion coefficient we have derived is, in fact, the eddy diffusion coefficient perpendicular to the mean mixing surfaces of the large-scale quasi-horizontal eddies. It may more appropriately, therefore, be termed a "quasi-vertical" eddy diffusion coefficient in order to distinguish it from the truly vertical coefficient.

6. Conclusion

Coincident observations of a layer of volcanic material and a sharp minimum in the vertical distribution of ozone at an altitude of about 20-21 km over Boulder, Colo. (40N), in March and April 1964, provide evidence of the quasi-horizontal advection of a thin and remarkably stable layer of tropical air into which volcanic debris was injected when Mt. Agung erupted on 17 March 1963.

Measurements indicate a peak particle concentration over Boulder of about one particle cm^{-3} between 20 and 21 km altitude, assuming a mean particle diameter of 1 μ.

Significant destruction of ozone by the volcanic material is not indicated.

The persistence of the ozone-depleted, dust-laden layer requires a "quasi-vertical" eddy diffusion coefficient, in the region of the ozone minimum, of the order of 2.5×10^2 cm^2 sec^{-1} or less. This result is specific to the space-time scale of the phenomenon described above and should not be applied uncritically to other stratospheric processes.

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REFERENCES

Anderson, K. A., 1965: Thin atmospheric layers of radioactive debris during September 1961 and 1962. *J. Geophys. Res.*, **70**, 1139-1153.

Bigg, E. K., 1964: Atmospheric stratification revealed by twilight scattering. *Tellus*, **16**, 76–83.

Bolin, B., 1964: Gross-atmospheric circulation as deduced from radioactive tracers. *Res. in Geophys.*, **2**, Cambridge, Mass. Institute of Technology Press, 479–508.

Brewer, A. W., and J. R. Milford, 1960; The Oxford-Kew ozone sonde. *Proc. Roy. Soc. London*, **A256**, 470–495.

Danielsen, E. F., 1959: The laminar structure of the atmosphere and its relation to the concept of a tropopause. *Arch. Meteor., Geophys., Biokl.*, **A11**, 293–332.

Dütsch, H. U., 1956: Das atmosphärische Ozon als Indikator für Strömungen in der Stratosphäre. *Arch. Meteor., Geophys., Biokl.*, **A9**, 87–119.

Dyer, A. J., and B. B. Hicks, 1965: Stratospheric transport of volcanic dust inferred from solar radiation measurements. *Nature*, **208**, 131–133.

Flowers, E. C., and H. J. Viebrock, 1965: Solar radiation: An anomalous decrease of direct solar radiation. *Science*, **148**, 493–494.

Harris, B., 1964: Volcanic particles in the stratosphere. *Aust. J. Phys.*, **17**, 472–479.

Hering, W. S., 1964: Ozonesonde observations over North America, Volume 1. Research Report AFCRL-64-30(I), Air Force Cambridge Research Laboratories, Bedford, Mass., 512 pp.

——, and T. R. Borden, Jr., 1964: Ozonesonde observations over North America, Volume 2. Environmental Research Papers No. 38, AFCRL-64-30(II), Air Force Cambridge Research Laboratories, Bedford, Mass., 280 pp.

——, and ——, 1965: Ozonesonde observations over North America, Volume 3. Environmental Research Papers No. 133, AFCRL-64-30(III), Air Force Cambridge Research Laboratories, Bedford, Mass., 265 pp.

Hering, W. S., and H. U. Dütsch, 1965: Comparison of chemiluminescent and electrochemical ozonesonde observations. *J. Geophys. Res.*, **70**, 5483–5490.

Junge, C. E., C. W. Chagnon and J. E. Manson, 1961: Stratospheric aerosols. *J. Meteor.*, **18**, 81–108.

Link, F., and L. Neuzil, 1965: Dioptric tables of the Earth's atmosphere. *Czech. Acad. Sci. Astronom. Inst. Publ.*, No. 50.

Meinel, M. P., and A. B. Meinel, 1963: Late twilight glow of the ash stratum from the eruption of Agung volcano. *Science*, **142**, 582–583.

——, and ——, 1964: Height of the glow stratum from the eruption of Agung on Bali. *Nature*, **201**, 657–658.

Mossop, S. C., 1964: Volcanic dust collected at an altitude of 20 km. *Nature*, **203**, 824–827.

Newell, R. E., 1963: Transfer through the tropopause and within the stratosphere. *Quart. J. R. Meteor. Soc.*, **89**, 167–204.

Newkirk, G., and J. A. Eddy, 1964: Light scattering by particles in the upper atmosphere. *J. Atmos. Sci.*, **21**, 35–60.

Pittock, A. B., 1963: Determinations of the vertical distribution of ozone by twilight balloon photometry. *J. Geophys. Res.*, **68**, 5143–5155.

——, 1965: Possible destruction of ozone by volcanic material at 50 mb. *Nature*, **207**, 182.

Reed, R. J., 1950: The role of vertical motions in ozone-weather relationships. *J. Meteor.*, **7**, 263–267.

——, and K. E. German, 1965: A contribution to the problem of stratospheric diffusion by large-scale mixing. *Mon. Wea. Rev.*, **93**, 313–321.

Volz, F. E., 1964: Twilight phenomena caused by the eruption of Agung volcano. *Science*, **144**, 1121–1122.

——, 1965: Note on the global variation of stratospheric turbidity since the eruption of Agung volcano. *Tellus*, **17**, 513–515.