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AN AIRCRAFT, AEROSOL SAMPLING PROGRAM: SOME PRELIMINARY RESULTS

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Abstract—An aircraft sampling program to determine the vertical and size distributions of that part of the atmospheric aerosol with radius greater than 0.25μ is described. The feasibility of using a single engine aircraft and a light scattering particle analyzer, for this work, is shown. Examples of the data are given and preliminary conclusions are drawn about the normal size distribution, effect of lapse rate on the vertical aerosol concentration gradient, effect of relative humidity on the size distribution, and the possibility of producing high concentrations of giant particles aloft.

1. INTRODUCTION

FOR A complete understanding of the effects of aerosols on the atmosphere, a knowledge of the vertical distribution and size distribution is required. The purpose of this paper is to describe an aircraft sampling program which is intended to obtain more information on these factors and to present some preliminary results which we feel are of interest.

The platform for these measurements is a single engine, low wing aircraft. Although there are serious questions about the applicability of single engine aircraft to this type of measurement, we believe we have shown that useful information can be obtained. The variables which are monitored are the ambient and dew point temperatures and the particle concentrations in two size ranges. To obtain a more continuous record of the aerosol concentrations than is available from batch sampling techniques, such as impaction or filter devices, a single particle, light scattering analyzer is used. Although these instruments do not usually measure the absolute size of the particles, we believe they are adequate for this work.

Other data which are presented are concerned with the vertical distribution and change in the size distribution of the aerosol with altitude. We have shown the effects of inversions on these factors and that changes in relative humidity at values as low as 50 per cent can cause measurable changes in the size distribution. Finally, we present evidence for the formation of giant particles aloft.

2. BACKGROUND

The following is a brief survey of the literature, which has appeared since 1967, concerned with the vertical and size distribution of the atmospheric aerosol in the lower troposphere. For a review of the literature prior to this, see DUFOR (1967).

The most common method of obtaining the number concentration and size distribution of an aerosol is to collect the particles with an impactor and then examine them microscopically. Aircraft soundings have been made using this technique by NAGY (1969) MEZZAROS, (1969), BLIFFORD and RINGER (1969) and BLIFFORD (1970). The results of these measurements showed that the exponent, β , in the log radius-number

distribution (JUNGE, 1963), $dN/d\log r = cr^{-\beta}$, varied from 2 to 3.6. Also, in general, the concentration decreased upward and reached values varying from 0.4 cm^{-3} at 9.1 km for particles $0.13 < r < 5.5 \mu$ (Blifford) to 0.01 cm^{-3} for particles greater than 0.5μ radius, above temperature inversions at 3 km (Nagy). Unfortunately, this is a very tedious and time consuming operation and because of the batch nature of the sampling does not allow a continuous determination on either a time or spatial basis.

Undoubtedly the most complete series of measurements on the vertical distribution of aerosols has been taken at three mountain stations at 700, 1800 and 3000 m by R. Reiter and his co-workers (REITER, CARNUTH and SLADKOVIC, 1968; REITER and SLADKOVIC, 1970; CARNUTH, 1970). These data, collected with impactors and a combination of diffusion boxes and a condensation nucleus counter, were analyzed with respect to air mass type, time of day, season, frontal passage, u.v. radiation, precipitation, cloudiness, and the mass exchange coefficient. Again it was found that the concentration of the aerosol particles decreased upward, temperature inversions hindered the upward transport and β varied from 2 to 4. Unfortunately, as these were surface measurements, there may have been unknown influences of the terrain.

Other measurement methods are based on various optical properties of the aerosol. One of the oldest methods to measure turbidity, the Volz sun photometer, was used by BACH (1971) in a polluted urban environment. He found, under clear skies, "enormously diverse stratification" in this atmosphere and that the lowest 1000 m could account for about 65 per cent of the total solar attenuation. Another optical instrument, the integrating nephelometer (e.g. CHARLSON, HORVATH and PUESCHEL, 1967; RAE and GARLAND, 1970), although larger and more cumbersome than the sun photometer, is also portable and allows more detail to be obtained on both a time and spatial basis than the sun photometer. In at least one case (AHLQUIST and CHARLSON, 1968) it has been carried in an aircraft. Although the data reported show more detail than that of BACH (above), there was undoubtedly more structure than was shown. Also, this method does not readily permit a determination of the size distribution.

Another method which shows great promise is lidar. While the majority of this research has been concerned with high altitudes, some measurements have been made in the troposphere (e.g. VIEZEE and OBLANAS, 1968; LANE *et al.*, 1971). Unfortunately, as with most optical methods, it is exceedingly difficult to invert the data to an accurate description of the aerosol because the measured signal is a function not only of the size and concentration of the particles present but of their shape and index of refraction as well. However, the published results do show the great variability that exists in the vertical, and horizontal, distributions.

The final method we would like to discuss is the use of single particle, light scattering analyzers. As this is the type of instrument we have used, we would like to review the literature in a little more detail.

These instruments measure the amount of light scattered from individual particles as they pass through an illuminated volume. Again the amount of scattered light will be a function of the refractive index, shape, and size of the particle, and as these factors are not known an absolute size determination cannot be made by this method. As monodisperse, polystyrene latex spheres of known radii are used in the calibration, the instrument gives only an equivalent size.

JUNGE (1966) suggests that because of the water soluble materials present in natura,

aerosols, even in clear air and particularly at the higher relative humidities, the particles will be approximately spherical with a refractive index close to 1.5. HUSAR *et al.* (1970) have shown that for one particular counter, the Royco PC220, if the refractive index is between 1.4 and 1.8, the ratio of the geometrical size to the size measured by the instrument, the "equivalent latex sphere" size, is between 0.9 and 1.5 with a "mean ratio being on the order of 1. . .". BLIFFORD (1970) showed that in the size range $0.5\text{--}5.0\ \mu$ the results from an optical counter agreed within an average factor of 1.6 with impactor results for sampling conducted at 8.5 km. HIDY *et al.* (1970) also suggest agreement within a factor of two between the results of their impactor samples and "those from the optical counters of Blifford and Ringer". Finally, JUNG, ROBINSON and LUDWIG (1969) have stated, "the absolute concentration may be somewhat in error . . . , but the general shape of the distribution is as good as the data obtained with most other methods".

Although a possible error of 100 per cent is usually not considered very good scientific accuracy, it may be justified in this work for the following reasons.

- (a) In actuality the real concentrations can vary by as much as two orders of magnitude so that an error of 100 per cent is relatively small.
- (b) Although the measured concentrations and even size distributions may be in error, changes of a factor of two in concentration or changes in the size distribution and properties of the aerosol, will be readily apparent. That is, even though the absolute numbers may be in error, changes should be obvious.
- (c) These instruments, with all their inaccuracies, appear to be the only ones which are capable of giving specific, although somewhat inaccurate, information on size distribution with a minimum of effort.

HIDY *et al.* (1970) performed a number of aircraft measurements over Northeastern Colorado using an optical counter, a CN counter, and filter and impactor samples. The results show concentrations of particles with radii greater than $0.15\ \mu$ of about $1\ \text{cm}^{-3}$ in relatively unpolluted areas. It is suggested that these concentrations are typical of the residual material in clean air and that higher concentrations are caused by local sources.

During a number of aircraft flights, measurements of aerosol concentration and size distribution in Florida and the Southwest, both in clean air and that contaminated with the efflux from power plants and forest fires, were made by MCCALDIN and JOHNSON (1969) and MCCALDIN, JOHNSON and STEPHENS (1969). These data indicate the possibility of tracking particulate pollution with optical scattering analyzers and show a "background" concentration of $1\text{--}3\ \text{cm}^{-3}$.

In tower measurements from 70, 170 and 500 ft, PETERSON, PAULUS and FOLEY (1969) have published data obtained in an urban environment using a CN and electrical counter, as well as an optical counter. The data which are reported were taken during times of negative, isothermal, positive, and superadiabatic lapse rates. Among the conclusions and hypotheses drawn from these data were:

- (1) a strong concentration gradient extends across the base of a temperature inversion;
 - (2) the average size above the inversion is larger than below;
 - (3) the variation in the average size with height is largest under inversion conditions;
- and

- (4) the average particle size is generally smaller under subadiabatic than inversion conditions.

Finally, mention might be made of the work previously quoted by JUNG (1966) and JUNG, ROBINSON and LUDWIG (1969), in which an optical particle analyzer was used to determine the size distribution of particles with radii greater than 0.15μ . From surface measurements made at Cape Blanco and Crater Lake, Oregon, three types of atmospheric aerosols were distinguishable. "The first, low-to mid-tropospheric aerosols, is characterized by size distributions that can be approximated by a power law with an exponent (β) of 3-4." The second type, which exists below about 2 km, is essentially the first type with the addition of components from sea spray. Finally, there were distributions representative of the high troposphere which were collected during periods of subsidence. In this latter case β took on a value of about 2.

On the basis of the above review, the following generalizations may be made.

- (1) The major source of the atmospheric aerosol is the earth's surface.
- (2) The vertical temperature structure is important in the dispersion of the aerosol.
- (3) Although wind induced mixing must also be important, little attention has been paid to this factor.
- (4) While the exponent, β , in the log radius-number distribution is not constant, it is often around 3. The actual value may indicate something about the previous history of the aerosol.
- (5) A "background" concentration of approximately 1 cm^{-3} of particles greater than 0.15μ radius may exist.
- (6) Single particle, optical scattering analyzers, although not completely accurate, are reasonable instruments for learning more about the atmospheric aerosol, its distribution and history.

3. EXPERIMENTAL APPARATUS AND DATA COLLECTION

3.1. Instrumentation and sampling procedure

Instruments to measure the ambient and dew point temperatures and concentration of particles in two size ranges are mounted in a single engine, Beechcraft Bonanza. A sampling tube, of 0.5 in. o.d. aluminum, 2 ft long, is attached to the air conditioner scoop on top of the fuselage. This tube is bent with two 45° , 7-in. radius bends so that it points along the longitudinal axis of the aircraft. The inlet is 8 in. above the fuselage and 10 ft behind and approximately in line with the top of the propeller arc. From the outlet of this tube, the air is carried by 3 ft of 0.5 in. tygon tubing into the cabin where it is exhausted. One end of a 2-in. length of 0.25-in. tygon tubing is inserted into this larger tubing while the other end leads directly to a 0.1 l. min^{-1} , Climet, model CI-201, particle analyzer. The output voltage pulses from the analyzer are fed to a Climet, model CI-205, 2-channel counter, in which the upper and lower limits of the pulses which are to be counted can be set for each channel. At the end of the specified count time, the totals in each channel are printed on a digital printer.

Temperature and dew point data are obtained with a Technology/Versatronics, Inc., Thermoelectric Dew Point Hygrometer. The ambient temperature is sensed with a thermistor which is attached 0.25 in. below and 0.75 in. behind the inlet of the sampling probe. For the dew point measurements, 0.8 l. min^{-1} of air is withdrawn

from the main sample line where it first enters the cabin and is passed, immediately, through the dew point sensor. The ambient and dew point temperatures are recorded on a strip chart recorder.

Sampling starts at the beginning of the take-off roll. As soon as practicable after take-off, the indicated airspeed is increased to 90 kts, and a climb rate of 150 m min^{-1} is maintained as long as possible. The climb is usually maintained to 3000 m or to such an altitude that the aerosol concentration is very low. At this point a descent is usually initiated, still at 90 kts and 150 m min^{-1} descent rate, and sampling continued until touchdown.

In general we have found that the most useful pulses to count are those which correspond to equivalent latex sphere radii of $0.25\text{--}0.5 \mu$ and $0.5\text{--}1.5 \mu$. Although the counter will accept pulses corresponding to an equivalent radius of 0.15μ , we question the accuracy in this range. Above 1.5μ , the concentration of particles is usually so low that excessively long counting times are required to obtain statistically meaningful results. With a count time of five seconds and a two and one half second hold time between successive counts, the particle concentrations are usually integrated over 13 m and recorded every 19 m.

3.2. Errors and calibrations

3.2.1. Aerosol concentration and size. The particle analyzer measures the amount of light, produced by an incandescent source, which is scattered in the near forward direction by particles as they pass singly through an illuminated volume. As noted earlier, the amount of light which will be scattered from each particle is a function of its shape and index of refraction as well as size. Thus, any size determination using such a light scattering instrument is accurate only for those particles used in the original calibration of the instrument. As it is impossible to calibrate with the many different and complex mixtures of aerosol particles which exist in the atmosphere, the primary calibration is usually performed with an easily obtainable monodisperse aerosol, i.e. polystyrene latex spheres. This primary calibration, which is performed by the manufacturer, allows only the determination of an equivalent, latex sphere size. (A brief check of the primary calibration, as well as a secondary calibration involving only the electronics, was made in this laboratory.) The errors involved in relating equivalent to real sizes are not as serious as they might first appear when it is realized that even when aerosol particles are examined microscopically, the definition of the sizes of irregularly shaped particles involves certain approximations and averaging. As final justification for the use of this type of instrument, we refer to the previously mentioned works of JUNGE (1966), JUNGE *et al.* (1969), BLIFFORD (1969), HIDY *et al.* (1970) and HUSAR *et al.* (1970).

The basic concern, therefore, is whether the instrument sees a sample representative of the air through which the aircraft is passing. Unfortunately, it is impossible to answer this question unequivocally as it is not possible to produce a known aerosol through which to fly. While other sampling techniques, such as impaction, could be used simultaneously to collect a sample of the same aerosol, there is no assurance there would not be errors in this sampling technique. Therefore, it is argued that the best way to check for possible sampling errors is to change the flight conditions to exaggerate those factors which may cause an unrepresentative sample to reach the

instrument. Any changes in measured concentrations with change in flight condition would be a strong indication of a sampling error.

Two of the major possible sources of error would be non-isokinetic sampling and loss of aerosol in the sample line. To check for these errors, the concentrations in various size ranges were determined at different airspeeds and angles of attack; i.e. in level flights, climbs, and descents. The results of these measurements, conducted in an area with relatively good spatial homogeneity of aerosol content, are shown in TABLE 1. In this and TABLE 2, the concentrations are listed in units relative to the data collected at 90 kts indicated airspeed (IAS), level flight and 2200 rev min⁻¹. R_c is the ratio of the concentrations of the smaller to larger sizes (see section 4). As the flowmeter in the analyzer is position sensitive, it is meaningless to try to maintain the same flow rate in different configurations or airspeeds. Thus, for these measurements the flow was set in level flight at 90 kts and not changed. As can be seen, there appears to be little correspondence between the various flight attitudes and airspeeds and the measured concentrations. At the higher airspeeds the measured concentrations may be slightly higher which is not unreasonable since no adjustments in the flow rates were made. The fact that the variation in R_c is less than ± 12 per cent is particularly good evidence that non-isokinetic sampling and impaction in the sampling system are not disqualifying factors.

TABLE 1.

IAS (kts)	Vertical speed (m min ⁻¹)	Relative particle concentrations		
		0.25-0.5 μ	0.5-1.5 μ	R_c
60	0	11.1	1.1	10.1
72	0	9.4	0.82	11.5
90	0	10.00	1.00	10.00
105	0	10.6	1.1	9.6
115	0	11.6	1.2	9.7
75	150 up	10.9	0.9	12.1
85	150 up	11.0	1.0	11.0
68	150 down	8.6	0.88	9.8
105	150 down	11.4	1.1	10.4

Another cause for concern is the inadvertent detection of the engine exhaust. Although the exhaust is emitted slightly ahead of, but below, the wing, other data show no difference between power operations and those in which the engine was completely shut down. Also, to determine whether aerosols produced in the cabin were reaching the analyzer, a large amount of smoke was produced. There was no increase in the measured concentrations at that time.

Other than affecting the trajectory of the sample air entering the sample probe, the propeller may have another effect. Although less than sonic, at high rev min⁻¹ there is cooling of the air at the propeller tips. This cooling can, particularly in regions of high relative humidity, cause condensation of water vapor on already existing aerosol particles. Although theoretical calculations of the cooling produced at the tips could be made, this alone would give little information about changes in the aerosol or

humidity of the sample air before it finally reaches the instruments. That this is not a serious problem was verified by operating in a region of relatively high relative humidity, 87 per cent, at constant altitude and airspeed (90 kts IAS) and different propeller rev min⁻¹. In TABLE 2 are shown the results of these measurements.

TABLE 2.

Rev min ⁻¹	Relative particle concentrations		
	0.25–0.5 μ	0.5–1.5 μ	R_c
1800	9.9	1.01	9.8
2000	9.8	0.97	10.1
2200	10.00	1.00	10.00
2350	10.1	1.03	9.8

While these tests do not prove that there are no errors in the sampling procedure, they do indicate that, if present, they are independent of the flight configuration. Thus, although the absolute values may be in error, differences are real.

3.2.2. Temperature. The system for measuring the ambient temperature was calibrated against a mercury-glass thermometer in the laboratory. From 2 to 34°C, the two agreed within $\pm 0.2^\circ\text{C}$ and the error was very reproducible. The effect of dynamic heating was determined by varying the IAS from 70 to 140 kts at constant altitude. The results showed that the response was very close to theoretical, and the average of four calibrations was

$$T_{\text{true}} = T_{\text{measured}} - 1.74 \times 10^{-4}(\text{IAS})^2.$$

In all data analyses, the ambient temperature is corrected for this dynamic heating. Attempts to determine the effects of radiational heating have been so inconclusive that we have felt the design, construction, and testing of a radiation shield was not worthwhile.

The dew point sensor was calibrated against a Cambridge, model 992, dew point hygrometer. In the dew point range 0–24°C, the errors varied between about -0.6 and $+0.3^\circ\text{C}$ with reproducibility no better than $\pm 0.2^\circ\text{C}$. As the Cambridge instrument is not accurate to better than a few tenths of a degree, no corrections are made to the measured dew points. Below 0°C, the errors were as much as $\pm 2^\circ\text{C}$ with very poor reproducibility and slow response.

Flight conditions should have no effect on the dew point of the air entering the sample tube unless there is condensation or evaporation on the aerosol particles. Although condensation and evaporation in the sample tube are possibilities, there is a high rate of air flow through this tubing and all but about 4 in. is metal. Flight evidence indicates that equilibration of the water vapor between sample air and tubing is no slower than the instrument response time which is specified by the manufacturer as "typically 5°F s⁻¹".

4. DATA AND DISCUSSION

Data from some of the more than 50 vertical soundings which have been made are shown in FIGS. 1-7. Unless noted otherwise, the parameters, which are plotted against altitude in meters, are:

T_A dry bulb temperature, °C.

RH relative humidity, per cent; calculated from dry bulb and dew point temperatures.

LPC concentration of large particles, $0.5 < r < 1.5 \mu$, number cm^{-3} .

R_c ratio of concentrations of small particles, $0.25 < r < 0.5 \mu$, to large particles.

To save space, the concentrations of small particles are not plotted. The large particle concentration curves have been smoothed to the extent of averaging every particle count with the adjacent one. Thus, there is a calculated value, the point

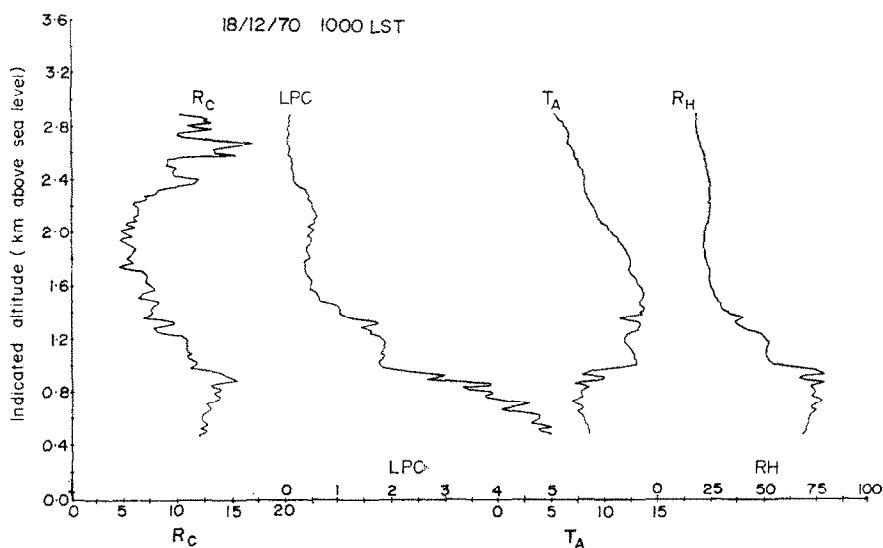


FIG. 1. 'Typical' winter sounding.

plotted, corresponding to every 7.5 s, approximately 20 m, which is the average of two counts taken over 15 s, approximately 40 m. The ratios were calculated from these average concentrations. In light of the previous comments regarding the calibration of the particle analyzer, no attempt has been made to assign possible errors to the plotted values of either the concentrations or ratios as we believe these would be largely meaningless. An indication of the reproducibility of the measurements may be obtained by noting that there are no wide, random fluctuations in the data as plotted. This is also true of the raw data.

FIGURES 1 and 2 are examples of fairly typical winter and summer data taken in relatively unpolluted conditions on 18 December 1970 near Rolla, Missouri, and 8 May 1971 near Springfield, Illinois. As would be expected, the winter sounding shows a marked temperature inversion and decrease in particle concentration above that level. Although not as obvious as in many soundings, immediately above the inversion the ratio of the concentrations also decreases. A decreasing ratio means that the

concentration of small particles is diminishing faster than that of the large particles. Whether this phenomenon is a result of the differing histories of the aerosol above and below the inversion or the usual concomitant change in the relative humidity is not known but we favor the latter explanation. Further evidence for this is contained in FIG. 2. Although this sounding is much different than the former one, with the exception of the spike at 3300 m, the correspondence between the ratio and relative humidity is quite apparent between 2000 and 3600 m. In this region, while the large particle concentration remains relatively constant, the ratio varies between 6 and 24 while the relative humidity varies between 40 and 66 per cent. These data also suggest that particle size is affected at relative humidities considerably lower than the approximately 70 per cent usually considered necessary for the interaction of the majority of naturally occurring aerosols and water vapor.

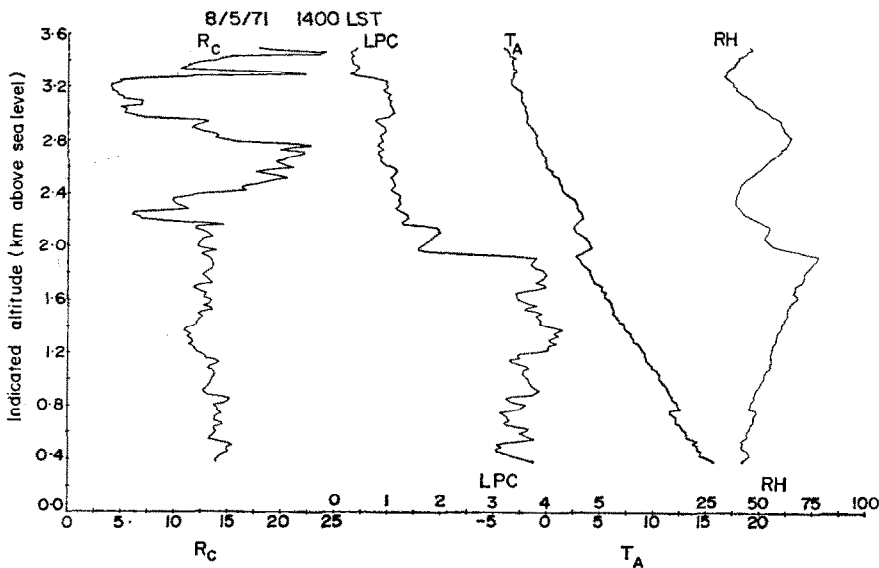


FIG. 2. 'Typical' summer sounding.

One of the primary interests in the value of the ratio is that, if the aerosol size distribution is correctly described by the log radius-number distribution, then R_c and β are related by

$$R_c = \frac{0.5^{-\beta} - 0.25^{-\beta}}{1.5^{-\beta} - 0.5^{-\beta}}.$$

TABLE 3 shows the values of β which correspond to various values of R_c .

TABLE 3.

R_c	β	R_c	β
0.98	0.5	10.5	3.5
1.5	1.0	15.2	4.0
2.3	1.5	21.8	4.5
3.4	2.0	31.1	5.0
5.0	2.5	63.1	6.0
7.3	3.0		

As exemplified in FIGS. 1-5, the most common values for the ratio correspond to β 's between 3 and 4. However, wide variations do occur and often for no apparent reasons. A prime example of this is shown in FIG. 3 which is data taken 28 November, 1970 approximately 45 miles southwest of Chattanooga, Tennessee. Up to 1600 m, while the concentration of large particles varied markedly, the concentration of small particles varied in the same manner so that the ratio was relatively constant between 13 and 14. At 1600 m, with no marked change in the large particle concentration, the ratio started to increase and reached a maximum of 28 at 2100 m and then decreased rapidly. While the decrease above 2100 m is not surprising in view of the marked decrease in humidity at this altitude, the reason for the increase between 1600 and

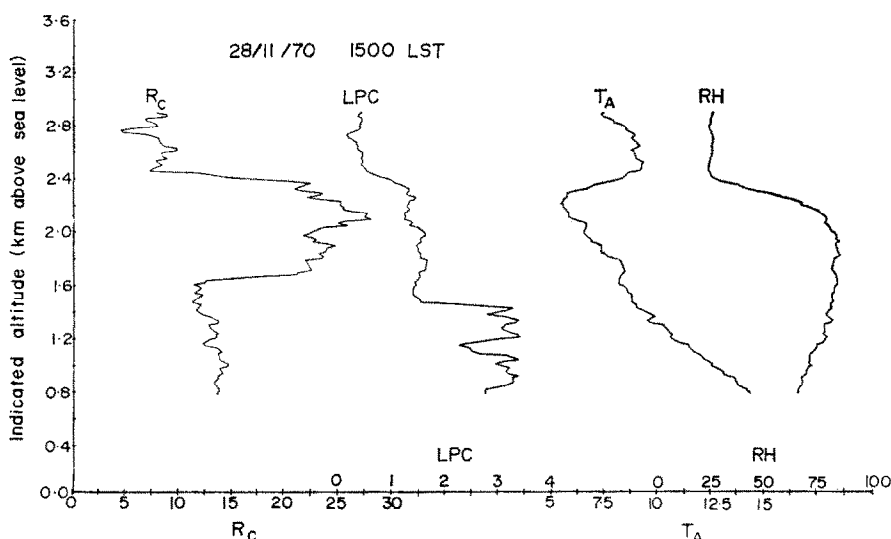


FIG. 3. Sounding showing blocking effect of small temperature inversion at 1600 m.

2100 m is not so obvious. From the data the only explanation for this distinct change in the distribution is the small temperature inversion which existed at 1600 m. Although this is not a pronounced inversion and would probably not have been identified in a regular radiosonde sounding, it must have significantly hindered the mixing of the aerosols so that two such distinct distributions could be formed.

The final results we would like to discuss concern the production of giant aerosol particles aloft. In FIG. 4 are shown the data obtained in a descent started at 1054 LST near Rolla, Missouri, 17 December, 1970. (Unfortunately, the relative humidity record was not complete for this sounding.) From 1800 to 2400 m there was a very large increase in the large particle concentration concurrent with a large decrease in the ratio. In this same region there was a noticeable odor reminiscent of an open sewer which we tentatively ascribe to H_2S . On the ascent just prior to this sounding, similar but much smaller changes in the large particle concentration and ratio were noted but the peculiar odor was not. During a subsequent ascent initiated at 1330 LST, the changes in the concentration and ratio were not found but a slight odor may have been detected at various altitudes between 1050 and 2050 m. It should be noted that the

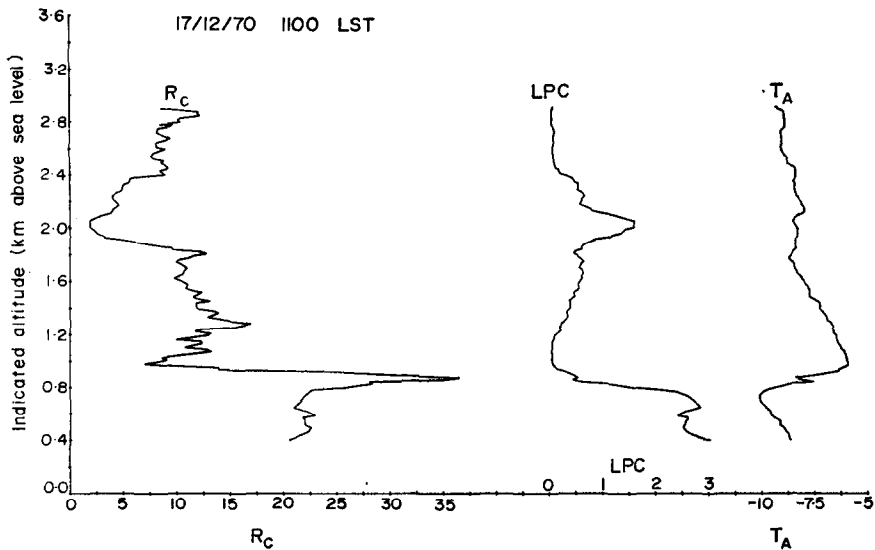


FIG. 4. Sounding which shows anomalously low R_c from 1800 to 2400 m.

overcast which had existed all the previous day had started to break at 1000 LST and it was clear by 1100.

Two much more pronounced examples of this phenomenon are shown in Figs. 5–7. The data in FIG. 5 were obtained in an ascent started at 0755 LST, 26 November, 1970, from Rolla. This ascent passed through one of a number of large holes which had appeared in a previously overcast sky. This thin cloud layer, which extended from approximately 850 to 1000 m, again covered the whole sky by 0930. This sounding appeared perfectly normal until a very strong temperature inversion was encountered, at which point the large particle concentration increased dramatically, the ratio fell to less than 1 and the same odor was very noticeable. As this was the first time these conditions had been encountered, their uniqueness was not realized and the sounding was not, unfortunately, continued.

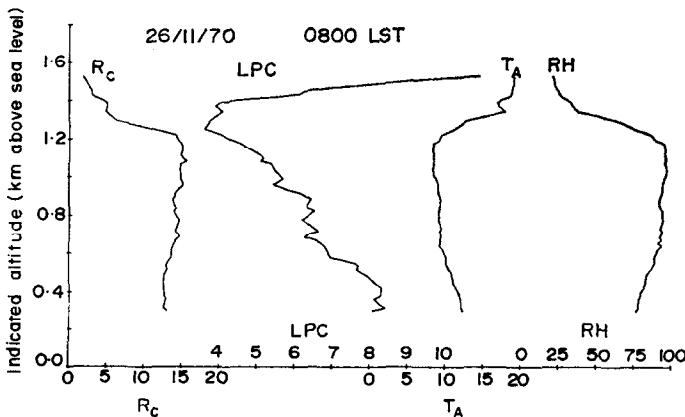


FIG. 5. Sounding which shows exceedingly high LPC and low R_c immediately above pronounced temperature inversion.

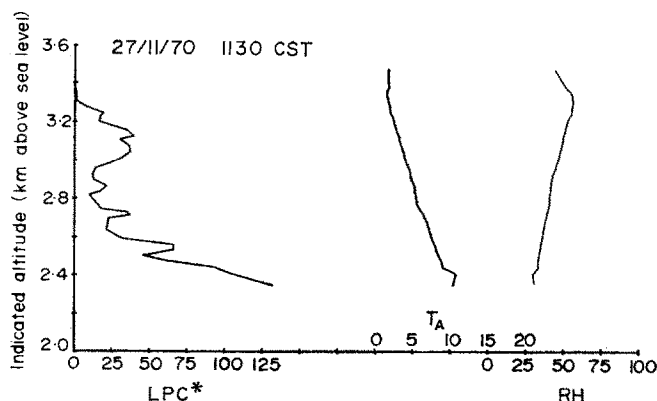


FIG. 6. Another example of high LPC and low R_c . Note: LPC* refers to particles with radius greater than 5μ .

One day later and about 150 miles to the southeast, the data in FIG. 6 were obtained (Note that in this case the ratio is not plotted as the concentration of large particles refers to those with radius greater than 5μ , 10μ dia. and thus the ratio cannot be related to β .) Again the sky was overcast but in this case the bases were very ragged, at approximately 1050 m, and the clouds were layered to approximately 2050 m. Although data were not taken in this cloudy region, because the temperature at 1000 m was 11.3°C and 10.3°C at 2350 m and because the tops of the overcast were quite smooth, it can be fairly assumed that a temperature inversion existed below 2350 m. Between the tops of the clouds and the beginning of the region of high large particle concentration, about 225 m, the air was very clear. At 2350 m, where the concentration of particles greater than 5μ radius exceeded 125 l^{-1} and the same peculiar odor was quite strong, the visibility was estimated at no more than 5 miles. FIGURE 7 is a histogram of the size distribution obtained at this altitude.

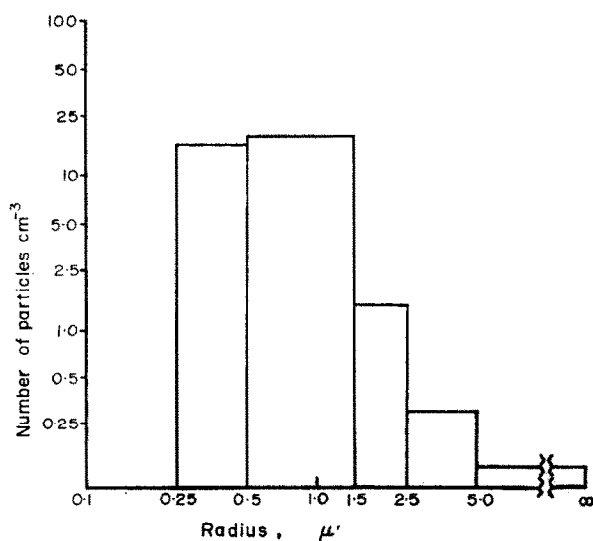


FIG. 7. Particle size distribution for aerosol at 2350 m for same sounding as FIG. 6.

Although we can offer no explanation for these cases of anomalously high large particle concentrations and low ratio, three factors stand out. In each case these unusual conditions appeared above a temperature inversion, and in the data of 17 December, 1970 considerably above, there was or had been an underlying cloud deck and there was always the peculiar odor. Because of the temperature inversions, it is unlikely that these particles could have been transported from the surface boundary layer at any time less than a number of hours preceeding the time that the measurements were made. Because of the height at which these anomalous conditions occurred or to which they extended, it is unlikely that these particles were the residues of evaporated cloud drops. While advection might explain high concentrations of particles, it does not explain why the ratio is so exceedingly low, lower than in any of our other measurements.

From the above arguments, we cannot help but believe that these particles were produced, or at least grew to these large sizes, through processes taking place above the surface of the earth. Further, because an odor reminiscent of H_2S was always present, it would appear that this substance is the cause, or result, of these processes. Unfortunately, neither gas nor aerosol samples were taken for laboratory analysis, and until such time as this is done we are handicapped in trying to postulate any specific mechanism to explain this phenomenon.

5. CONCLUSIONS

Although we have covered a number of different topics above, we would like to emphasize the fact that, at least in this work, a single engine aircraft proved feasible for aerosol sampling. Also, while single particle, light scattering analyzers do not measure absolute sizes, they are adequate for atmospheric sampling particularly when changes in concentration or size are of interest. They also allow greater detail on a time or spatial basis than is possible with other methods.

In this work more than fifty vertical soundings have been made. With so much data, no two sets of which are identical, there is a problem as how best to present the results. One possibility would be to combine the data from the various soundings and obtain an average of the various parameters as a function of altitude. However, as we have tried to show, small variations in the temperature gradient or relative humidity can cause large variations in the particle concentrations and size distributions so that averages are largely meaningless. Further, it is primarily these interrelationships in which we are interested. Thus, it did not appear worthwhile, at this time, to compute and graph such averages. If more data from a single location and taken on a more routine basis become available, then average profiles might serve a useful function as "benchmark" measurements.

In light of the above, we have shown only examples of the data which we believe illustrate certain behavior of the atmospheric aerosol. In the future we hope to be able to treat each of these topics individually. However, for the moment we would like only to present the following conclusions, some of which are tentative.

(1) Although, as commonly realized, the concentration of aerosols generally decreases upwards, when examined in detail this average behavior covers many instances of increasing concentration with altitude within distinct horizontal strata, these strata being defined, at least in part, by the temperature gradient.

(2) The most common value of β , the exponent in the log radius-number distribution, for particles between 0.25 and 1.5 μ radius, lies between 3 and 4 although values greater than 6 and smaller than 0.5 have been found.

(3) Changes in the water vapor content, at values as low as 50 per cent relative humidity, changes the relative concentrations of small particles, $0.25 < r < 0.5 \mu$, much more than large particles, $0.5 < r < 1.5 \mu$.

(4) Relatively high concentrations of giant particles can be produced aloft.

In summary, we would simply like to stress that the vertical and size distributions of the atmospheric aerosol are determined, in large part, by the fine structure of the atmospheric temperature and humidity gradients.

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