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SULFATES AND OTHER WATER SOLUBLES LARGER THAN 0.15 RADIUS IN A CONTINENTAL NONURBAN ATMOSPHERE

By

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ABSTRACT

Number concentrations of large and giant atmospheric particles and particles containing sulfate and water-soluble constituents during 10 days in March, 1969, were determined. Particles were collected by means of an Andersen multistage impactor and examined by means of an optical microscope. The number of particles collected and concentration of sulfate and water-soluble particles at the isolated New Mexico sampling site were comparable to literature-cited values of average continental concentrations over mountains or unpolluted areas. The number concentrations of giant and large particles did not appear to be influenced in the same way by meteorological parameters. Increases in the number of large particles were mirrored by corresponding increases in sulfate content.

Data for relating Andersen sampler aerosol number concentrations to concentrations reflected by the Royco 202 light scattering aerosol counter are given.

CONTENTS

INTRODUCTION

High concentrations of ammonium sulfate and sulfuric acid in various states of neutralization have been detected in major cities of the United States. Although it has been concluded that these chemical species are likely to be found near cities where sulfur dioxide effluent from industrial and residential heating abounds [1], ammonium sulfate particles and sulfate compounds have also been found off the coast of a nonindustrial portion of California [2]. Similarly, the infrared detection of ammonium sulfate [3] in the relatively clean atmosphere near White Sands Missile Range has suggested that even in an unpolluted area, sulfates may be a dominant species in the large particle size aerosol fraction (0.1 to 1.0 μ radius).

Since ammonium sulfate and sulfuric acid neutralization products are hygroscopic, they condense water from the air at moderately high relative humidities. The associated fog or haze and subsequent deterioration in visibility are of interest here from the standpoint of warm fog and haze dispersal or creation.

The determination of the size and number of sulfate and other water-soluble particles which could be a prime cause of the development and persistence of continental fog and haze in an unpolluted area has fostered this examination of individual particles present in the natural aerosol.

EXPERIMENTAL METHODS

To obtain a size-fractionated dust sample which could be analyzed chemically for sulfates, an Andersen sampler* was employed. The sampler is a multistage impactor [4] which was fitted with 8.26 cm diameter glass slides. Simultaneous to this study a Royco Particle Counter** was used to monitor continuously the number of giant particles at the same location [5].

Prior to sampling, the Andersen slides were coated with a transparent plastic which contained barium chloride as a sulfate-detecting reagent [6]. After exposure of the collected particles to water-saturated air, the sulfate particles reacted to form Liesegang rings of barium sulfate particles which were easily discernible microscopically. Since water-soluble components which serve as fog nucleants dissolve into the moistened plastic, further information on the number of potential fog or cloud droplet-forming nuclei was obtained.

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*Model 705, Medi-Comp Res. and Dev., Salt Lake City, Utah 84115.

**Model 202, Royco Instruments, Inc., Menlo Park, Calif. 94025

The size ranges which can be fractionated by the Andersen sampler are indicated in Table I. Size limits for each stage and for a unit density particle have been published [7]. The 95% limits (size limits which include 95% of the particles on a stage) for particles having a density of 1.77 gm cm^{-3} (ammonium sulfate density) and 2.4 gm cm^{-3} (estimated density of total particles captured) have been calculated using a conversion formula [7].

The sampling was carried out at Mule Peak [3] on a mountain range 1250 m above an adjoining desert floor in south-central New Mexico. The site was chosen because of its remoteness from anthropogenic particle sources and could yield aerosol data comparable to the number and chemical constituency of particles at other unpolluted locations. The Andersen sampler monitored air 9 m above ground, and the Royco sampled air from approximately the same height but was displaced 7 m ESE of the Andersen unit.

The particles were sampled with the Andersen instrument between 0200 and 0500 hours MST because the dust sampled at this time usually typified that of the boundary layer over the mountain on which the sampler was located better than the dust of any other of these daily periods [8]. The sampler was operated for 10 days in March, 1969, during the above-indicated 3-hour periods.

The meteorological conditions during the 10-day sampling period have been documented and analyzed [5]. The following parameters were measured or estimated: percent cloud cover, visibility, wind speed, wind direction, and relative humidity over the sampling site and adjacent basin.

The number of particles cm^{-3}, number of gm sulfate, and percent particles containing sulfate from each of the six Andersen sampler slides are given in Table II. A dash in place on an entry indicates number of particles or percent sulfate below the detection level (0.001 cm^{-3} and 0.1%). Each slide was examined before and after treating with water-saturated air to detect the disappearance of water-soluble components.

The giant particle concentrations are plotted in Figure 1. The continuous Royco data were averaged over the period of Andersen sampler operation to obtain one value per day; these compared well with Andersen data except for the concentrations of the 22nd.

The total number of particles cm^{-3} of giant and large particles (stages 1-4, 5 and 6 of the Andersen sampler) are plotted in the lower portion of Figure 2. A graph of percent particles containing sulfate is shown in the upper portion of Figure 2.

Table 1. Radius (μ) of 95% of Particles which can be Collected on Individual Stages of Andersen Sampler

Stage	1	2	3	4	5	6
Unit density [7]	>4.1	2.5 -5.25	1.5 -2.9	1.0 -1.75	0.4 -1.1	0.15-0.5
1.77 gm cm^{-3} density	>3.04	1.84-3.91	1.04-2.14	0.71-1.28	0.26-0.79	0.08-0.34
2.4 gm cm^{-3} density	>2.61	1.57-3.35	0.93-1.83	0.61-1.09	0.22-0.67	0.065-0.29

Table 11. Number concentrations of total particles, sulfate particles and percent particles containing sulfate

Stage (Andersen Sampler)	Total Particles No. cm^{-3}	SO_4, $\mu gm\ m^{-3}$	% Particles Containing SO_4	Stage (Andersen Sampler)	Total Particles No. cm^{-3}	SO_4, $\mu gm\ m^{-3}$	% Particles Containing SO_4
19 March				**22 March**			
1	0.017	0.03	4.3	1	----	----	----
2	0.01	0.02	1.4	2	0.05	0.02	0.25
3	0.02	0.02	6.2	3	0.07	0.01	0.45
4	0.04	0.01	5.0	4	0.15	0.00	0.25
***5	1.0	0.24	36.8	5	0.7	0.24	28.9
***6	90-165*	4.4	60.**	6	2.7	0.03	13.9
total		4.72		total		0.30	
20 March				**24 March**			
1	----	----	----	1	----	----	----
2	0.02	0.47	22.5	2	0.02	0.06	2.51
3	0.06	0.30	25.1	3	0.10	0.03	2.0
4	0.29	0.22	23	4	0.16	0.04	5.55
5	1.3	0.09	6.3	5	0.5	0.03	5.00
6	3.2	0.03	11.5	6	0.7	0.01	4.66
total		1.11		total		0.17	
21 March				**25 March**			
1	----	----	----	1	----	----	----
2	0.02	0.01	0.36	2	0.01	0.02	2.7
3	0.09	0.00	0.14	3	0.03	0.02	3.9
4	0.18	0.00	0.15	4	0.04	0.00	2.3
5	1.1	0.3	30.5	5	0.3	0.04	12.6
6	40-70*	1.6	50.2**	6	0.8	0.01	2.16
total		1.91		total		0.09	

*Estimated from aggregate reaction ring [6], stage size limits, and capture efficiency corrections.
**Estimated percent of reaction. Particles too numerous to count.
***Values of particle number and sulfate mass obtained from stages 5 and 6 have been corrected as stated under CALCULATIONS.

Table II (Continued)

Stage (Andersen Sampler)	Total Particles No. cm^{-3}	SO_4, $\mu gm\ m^{-3}$	% Particles Containing SO_4	Total Particles No. cm^{-3}	SO_4, $\mu gm\ m^{-3}$	% Particles Containing SO_4
26 March				**29 March**		
1	0.01	0.01	1.6	1	0.00	0.00	1.2
2	0.02	0.02	1.6	2	0.00	0.00	0
3	0.04	0.02	2.5	3	0.01	0.01	2.9
4	0.20	0.00	0.6	4	0.03	0.00	2.7
5	1.0	0.06	5.9	5	16	7.1	58
6	2.8	0.00	4.3	6	208-375*	12.6	75**
total		0.11		total		19.71	
27 March							
1	0.00	0.0	0				
2	----	0.0	----				
3	0.01	0.0	2.1				
4	0.02	0.0	0.9				
5	0.2	0.02	7.6				
6	0.8	0.01	6.8				
total		0.03					
28 March							
1	----	----	----				
2	0 00	0.00	1.8				
3	0.01	0.01	2.2				
4	0.02	0.00	2.5				
5	0.3	0.01	4.6				
6	>4.1*	1.4	10.8				
total		1.47					

*Estimated from aggregate reaction ring [6], stage size limits, and capture efficiency corrections.
**Estimated percent of reaction. Particles too numerous to count.

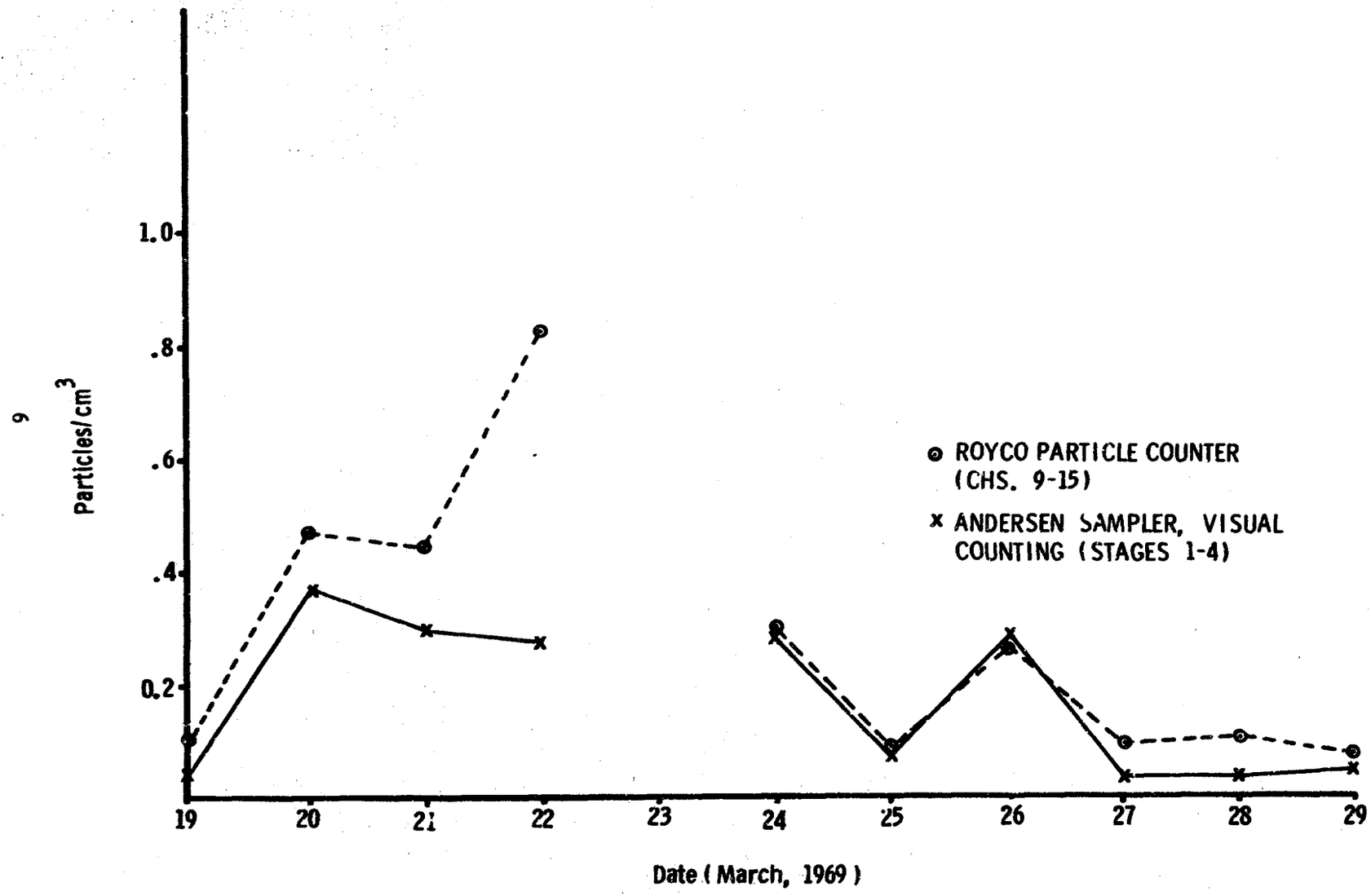


FIGURE 1. Comparison of giant particle number concentrations at Mule Peak, March, 1969.

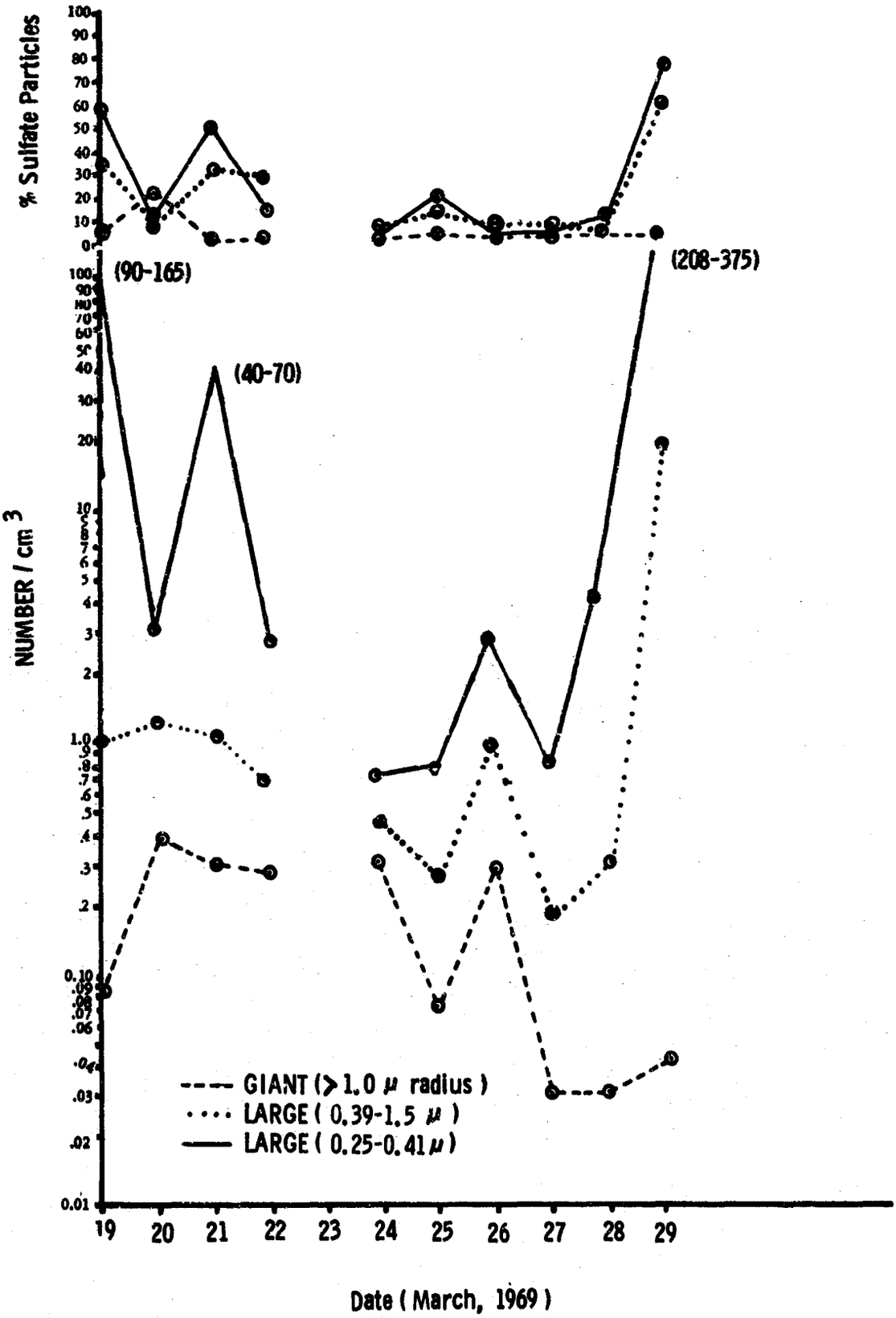


FIGURE 2. Number concentrations and percent sulfate in aerosol samples at Mule Peak, March, 1969.

DISCUSSION

A rather small number of samples have been examined; however, it appears that within the experimental period, the data allow certain inferences to be made with respect to particle number and constituency in the sampling region.

Giant Particles

Examination of percent cloud cover, visibility, wind speed, wind direction, and relative humidity over the site and adjacent basin revealed a primary dependency of giant particle number on wind speed in the adjoining basin floor, wind direction, frontal passage, history of air mass, and snow scavenging [5].

Except on the 22nd, the percent of giant particles containing sulfate (Figure 2) was below 5%. The air sample containing the high percentage (23.5%) of giant sulfate particles deserves comment because usually the continental large particle fraction contains the bulk of sulfate particles [9]. For this reason it appears that the sulfate particles sampled on the 22nd were picked up as giant particles rather than "growing up" from agglomerations of smaller sulfate particles which can result from the conversion of sulfur dioxide to sulfate [10].

Particle number concentrations and aerosol sulfate concentrations at locations comparable to Mule Peak are presented in Figures 3 and 4. Some of the values were calculated for ammonium sulfate from sulfur content. The points have been connected to make reading easier and, possibly they indicate the extent of anthropogenic influence.

The Mule Peak concentration of less than 0.05 $\mu gm\ m^{-3}$ sulfate in the giant size is less than most other continental observations, however, at times concentrations at Mule Peak were considerably higher (Table II). The low relative humidity usually present in the Mule Peak area may be influential in preventing the growth of large-sized hygroscopic sulfates into the giant range, a phenomenon which has been observed at other locations [11]. At Mule Peak, Budapest Observatory, off the California coast, in U. S. nonurban areas, etc. (Figure 4), where one might expect to find clean air, the values for sulfate concentrations are approximately 10 to 60 times the Izaña [12] value (during an influx of air from the Sahara). The high values at the other sites in Figure 4 when compared to the extremely low value at Izaña may indicate the extent of anthropogenic influence.

Percent sulfate mass in the giant size was 0.3% at Mule Peak which is consistent with the values of 1.4% and 1 to 2% near Budapest, Hungary, [20] for particles greater than 0.3 μ radius.

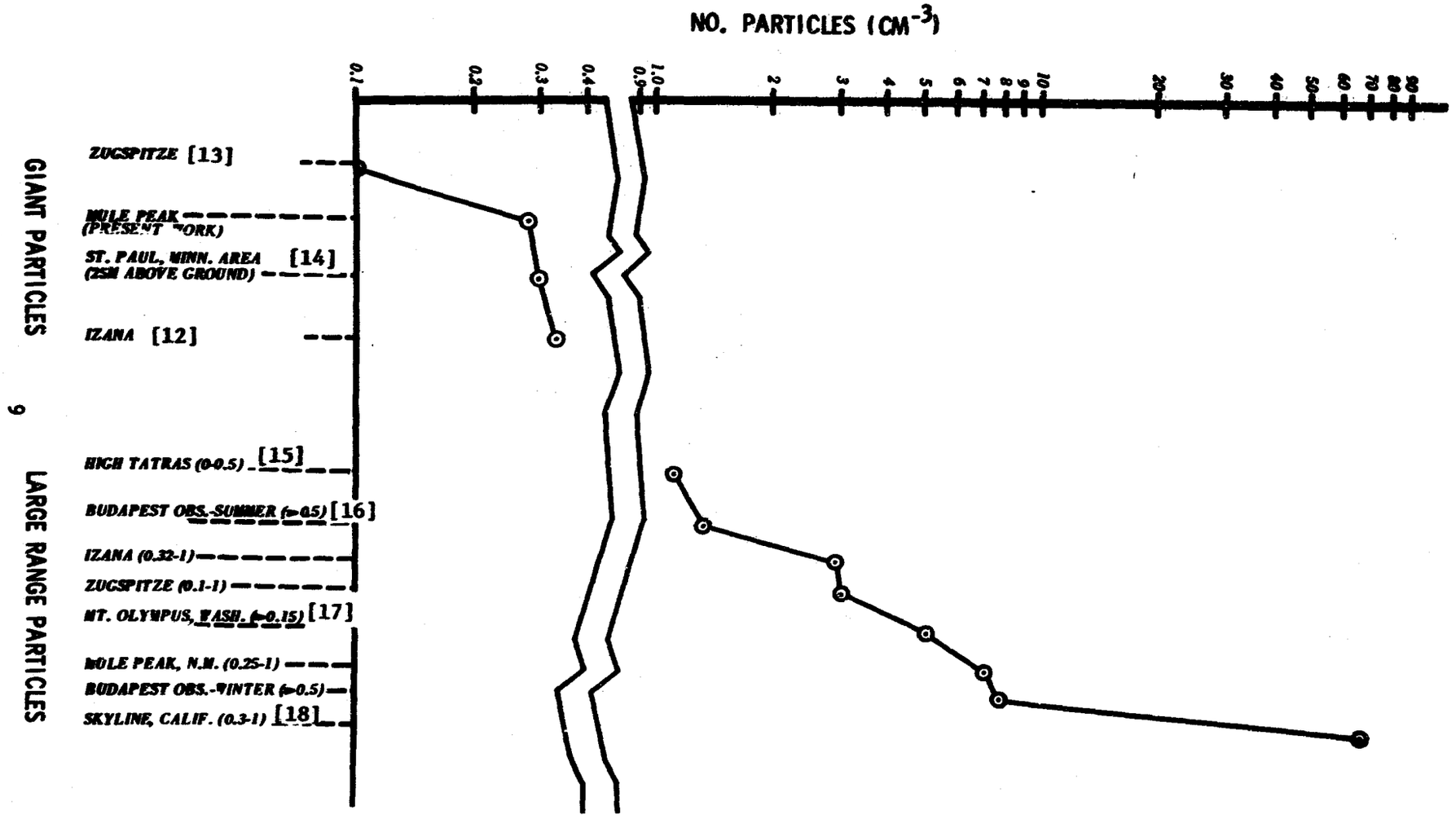
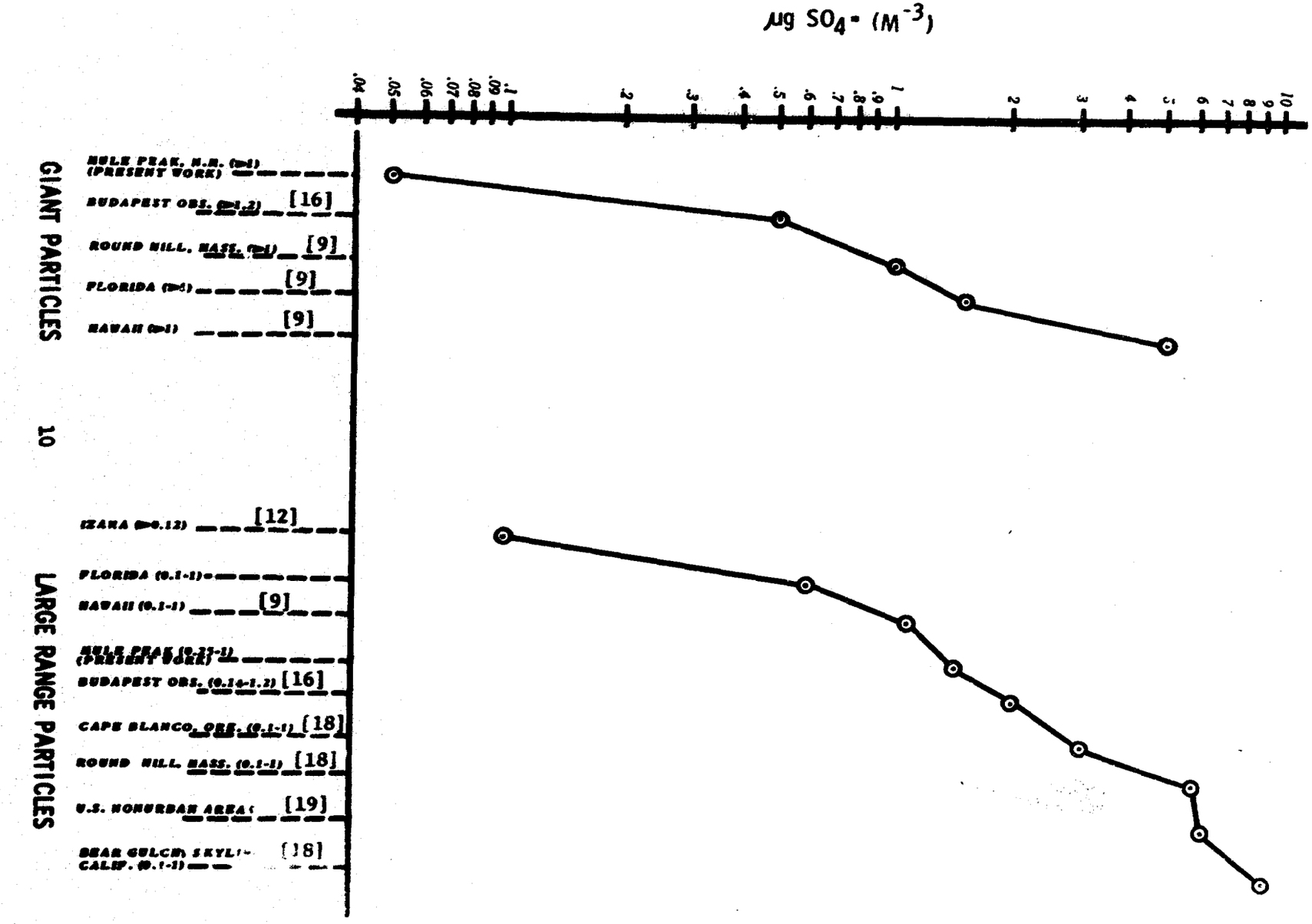


FIGURE 3. Comparison of particle concentration in mountainous areas.


μg $SO_4^=$ (M^{-3})
.04
.05
.06
.07
.08
.09
.1
.2
.3
.4
.5
.6
.7
.8
.9
1
2
3
4
5
6
7
8
9
10
GIANT PARTICLES
10
LARGE RANGE PARTICLES
MULE PEAK, N.H. (>1) (PRESENT WORK)
BUDAPEST OBS. (>1.2) [16]
ROUND HILL, MASS. (>1) [9]
FLORIDA (>1) [9]
HAWAII (>1) [9]
IZANA (>0.12) [12]
FLORIDA (0.1-1) =
HAWAII (0.1-1) [9]
MULE PEAK (0.23-1) (PRESENT WORK)
BUDAPEST OBS. (0.1&-1.2) [16]
CAPE BLANCO, ORE. (0.1-1) [18]
ROUND HILL, MASS. (0.1-1) [18]
U.S. NONURBAN AREA [19]
BEAR GULCH, SKYLIGHT CALIF. (0.1-1) [18]

Large Particles

It is apparent from Figure 2 that factors influencing the increase or decrease of giant particle number and sulfate concentration did not affect the same quantities in large particles in the period of observation. For instance, on the 21st, 25th, 28th, and 29th the number and percent sulfate for large particles rose; but giant particle numbers did not experience increases. Conversely, on the 20th the number concentration and percent sulfate rose for giant particles; but both these quantities decreased in the large particle size.

The change in number concentration of large particles was examined with respect to the previously mentioned meteorological parameters. Large particle sulfate and number concentration increased when the wind flow was over the mountain range on which the sampler was located and did not appear to be related to any other measured parameter. The difficulty in relating particle number and sulfate concentration to meteorological patterns has been experienced by others [21, 22].

Although Mule Peak large particle sulfate concentrations were at times high (Table II), the average value was approximately 1.4 μgm m^{-3}, which is similar to concentrations in Florida, Hawaii, and Budapest (Figure 3).

Soluble Particles

Except for the 22nd of March, the number of particles containing soluble nonsulfate was always less than 3.5% of the total particulate number. Percent nonsulfate solubles added to percent soluble sulfates gave a mode value of less than 10%, which falls short of Junge's estimate of soluble aerosol particles of 20 to 30% [9], and values of 15% in Los Angeles [23]. Data on the percent of water-soluble constituents in aerosol samples at comparable locations are scarce; however, the values of 7.5% and 10% near Budapest and in the country [24], given for the sum of soluble sulfate and ammonium chloride, can be examined. It should be noted that the Mule Peak value of 10% indicates percentage of particles containing solubles, and is not equivalent to the Bonis [24] percentage figures which compare the mass of chloride and sulfate to the total mass of collected particles.

Modal number concentrations of soluble giant particles at Mule Peak were comparable to those of Delhi, India [25]: Mule Peak mode 0.0015 cm^{-3}, maximum 0.09 cm^{-3}, Delhi 0.001 and 0.007.

As observed by others [20, 9], the soluble particles detected in the present study in the microscopic range were mixed in character. Both the data from Mule Peak and that from the Budapest Observatory [26] are in agreement in that the water-soluble fraction accounts for only a small part of the total aerosol mass.

CALCULATIONS

Data are presented here for determining correction factors to relate the particle counts on stages 5 and 6 of the Andersen sampler to the atmospheric aerosol number concentration as determined by a Royco model 202 particle counter.

Among other things, both the Andersen and Royco size classifications are based on the assumption that the particles are spherical. The size of a particle as judged by the Royco is mainly related to its size and refractive index [27]. In the Andersen, however, a particle is size-fractionated depending on its density. Thus a particle of size 1 μ and density 2.4 gm cm^{-3} behaves as an aerodynamically equivalent particle of 1.45 μ and unit density in the Andersen sampler.

Table III allows one to parallel channels in the Royco to the stages of the Andersen according to particle size employing densities between 1.7 and 2.4 gm cm^{-3}.

In an experiment designed to match Royco counts and Andersen sample counts the Andersen and Royco instruments were run simultaneously. Royco channels 1 and 2 were plotted against Andersen stage 6; Royco channels 3 through 8 were plotted against Andersen channel 5, resulting in the data presented in Figure 5. Royco channels above 8 (particles > 1 μ radius) showed fairly good correspondence to stage 4 (Figure 1); therefore, Andersen stage 4 counts were not changed.

Although Royco channels 6-8 are listed in Table II as being within stage 4 limits rather than stage 5 limits, the capture efficiency for these sizes on stage 4 is poor [7]. Similarly some of the particles which are in the 95% limits of stage 5 are captured on stage 4. This "trade off" of Royco channel counts between stages 4 and 5 of the Andersen seems to be useful in that stage 4 counts need no correction and stage 5 and 6 corrections can be obtained from the relationship given in Figure 5. This type of correction is not as applicable to chemical determination because chemistry of stage 4 and 5 particles may be different.

Andersen sampler number concentrations and percent sulfate values presented in Table II were corrected by means of the graph of Figure 5.

Uncorrected Andersen sampler number concentration values greater than 2.5 cm^{-3} were obtained from four slides on which there was an aggregate reaction [6]. From the graph of Figure 5 the largest uncorrected Andersen number concentrations required a correction of approximately four times the Andersen determination to match an equivalent Royco count. Because of the lack of points from which to make interpolations with confidence, number concentrations greater than 2.5 cm^{-3} were

Table III. Relationship Between Andersen Sampler and Royco Counter Size Ranges

Royco Ch. No.	Royco Radius Measured (μ)	Equivalent Radius (μ) for Andersen Sampler	Andersen Stage	
1	0.15-0.2	0.25- .38	5,6	6
2	0.2 -0.25	0.32- .41	5,6	
3	0.25-0.3	0.39- .41	5	5
4	0.3 -0.4	0.46- .69	4,5	
5	0.4 -0.5	0.60- .82	4,5	
6	0.5 -0.6	0.82- .90	4	
7	0.6 -0.75	0.90-1.10	4	
8	0.75-1.0	1.1 -1.50	4	

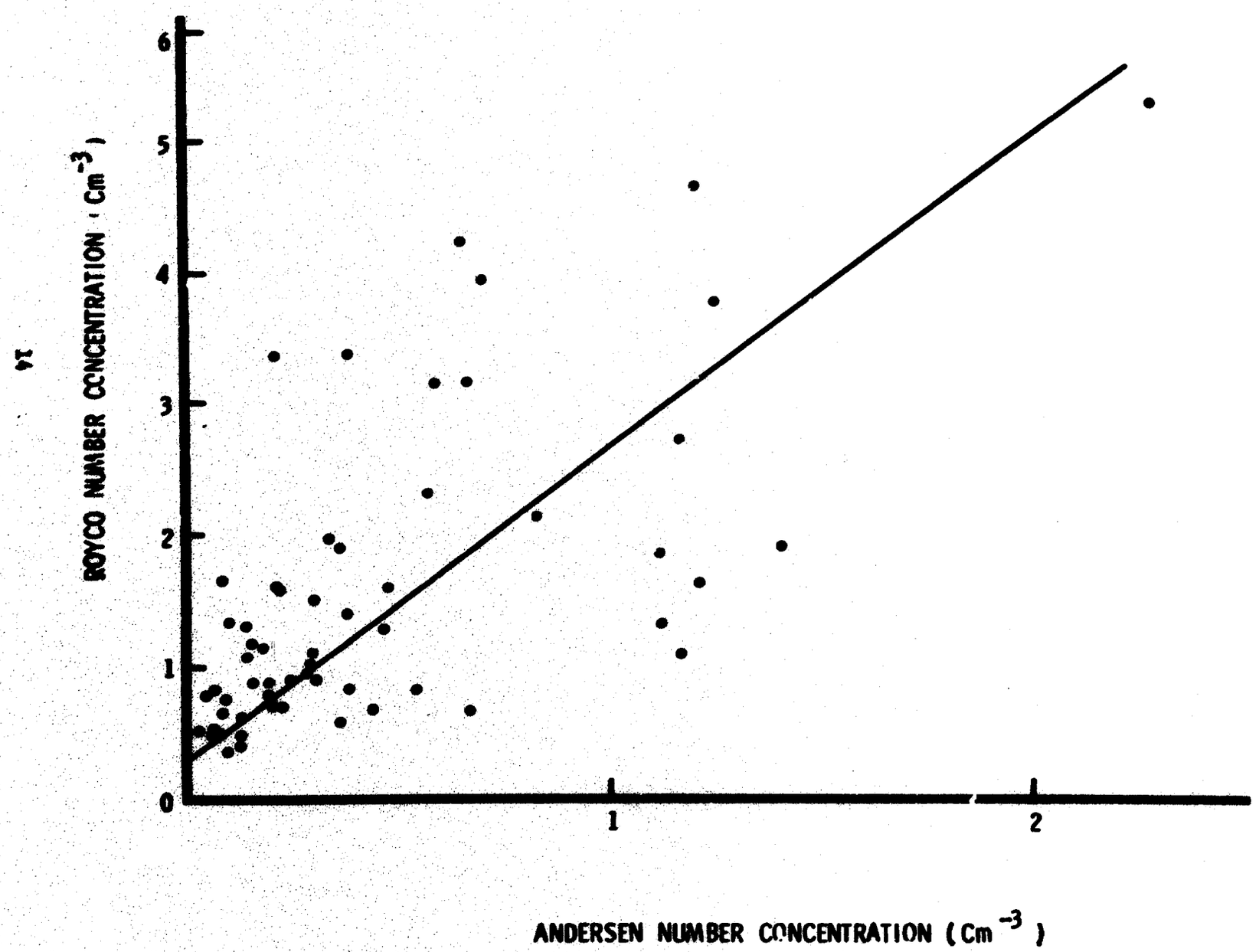


FIGURE 5. Relationship between Royco and Andersen instruments - large particle number concentrations.

simply multiplied by four to obtain an estimate of number concentrations of aerosol particles.

In general, Andersen values were lower than Royco values of aerosol number concentration by a factor of from 4 to 6. This difference is to be expected due to the rather poor efficiencies [7] on the Andersen sampler stages near the lower end of each stage size interval. The particle loss is augmented because as particle size decreases in the natural aerosol the number concentration of aerosol particles increases thereby causing the greatest number of particles at the lower limit of each slide.

Sulfate mass concentrations as ammonium sulfate were calculated by employing the corrected number concentrations along with percent particles containing sulfate values (Table I). The sulfate mass thus computed will represent an upper limit because most of the particles containing sulfate also contained a nonsulfate fraction.

CONCLUDING REMARKS

Pertinent to period and area of study, the following remarks concerning the data are made:

(1) Meteorological parameters impose a different effect on the fluctuations in giant and large particle sulfate content and number concentration (i.e., during the same period number of large particles increased while number of giant particles decreased).

(2) When there is a rise in number of large particles there is usually an increase in the percent of large particles containing sulfate.

(3) The giant particles are almost exclusively "mixed" in nature, that is, they are not entirely soluble, but contain an insoluble fraction.

(4) Individual large-sized particles contain a higher percentage sulfate per particle than giant particles and in many cases appear to be entirely sulfate.

(5) The soluble content of the aerosol samples (in the size range collected) is usually less than 10% of the total mass.

(6) The number of particles, soluble particle content, and sulfate concentrations of Mule Peak aerosol were comparable to results obtained at other mountain locations or rural locations.

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13. ABSTRACT

Number concentrations of large and giant atmospheric particles and particles containing sulfate and water-soluble constituents during 10 days in March, 1969, were determined. Particles were collected by means of an Andersen multistage impactor and examined by means of an optical microscope. The number of particles collected and concentration of sulfate and water-soluble particles at the isolated New Mexico sampling site were comparable to literature-cited values of average continental concentrations over mountains or unpolluted areas. The number concentrations of giant and large particles did not appear to be influenced in the same way by meteorological parameters. Increases in the number of large particles were mirrored by corresponding increases in sulfate content.

Data for relating Andersen sampler aerosol number concentrations to concentrations reflected by the Royco 202 light scattering aerosol counter are given.

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