Observations of aerosols and droplets in California stratus II

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ABSTRACT

Size distributions of aerosols and droplets were observed in stratus in the hills south of San Francisco during the summers of 1968 and 1969. Instrumentation included a Royco light-scattering particle counter, a condensation nuclei counter, and a settling tunnel. The average size distributions showed primary modes at submicron sizes on clear days, during cloud formation and dissipation, and in the developed cloud. A broad secondary mode of about 3 droplets $cm^{-3}\mu^{-1}$ was found at a radius of 5 μ in the cloud. A somewhat sharper mode of 0.9 $cm^{-3}\mu^{-1}$ at 8-μ radius was found in the average size distribution for dissipating clouds. During periods of cloud onset, the secondary mode is not well developed. Timesection analyses of particle and droplet spectra are given, and these show that there is considerable temporal variability in the spectra. The time-section analyses fail to confirm the general trend of the droplet mode toward larger sizes with time, as predicted by some numerical calculations of condensation processes. Analyses of the chloride, sulfur compound, and nitrate content of the aerosols and droplets are presented. The amounts of sulfur compounds and nitrates relative to chlorides are too high for the principal source of these materials to have been sea salt, unless the chlorides had been lost through some chemical process.

Introduction

The results presented here are an extension of work reported earlier (Ludwig & Robinson, 1970). The earlier results were based on observations made during the summer of 1968; this paper adds results obtained during the summer of 1969. The earlier paper was concerned with average aerosol and droplet size distributions; this paper presents examples of the changes in size distribution as a function of time. Finally, the previous paper presented quantitative measurements of the sulfur and chloride compounds in the aerosols and droplets; the present work also includes nitrate analyses.

Sampling instrumentation and procedures

The instrumentation and procedures used on this program to obtain particle and droplet size distributions have already been described in detail (Ludwig & Robinson, 1969, 1970) and will only be reviewed here very briefly. Except as noted, all the results presented in this paper came from the site referred to as "skyline" in previous work. This site is about 40 km south-southeast of San Francisco at the crest of the hills with an elevation of about 300 m. It is about 8 km east of the Pacific coast with very few pollution sources in that direction. Virtually all sampling was done with winds from the westerly quadrant.

Sampling instrumentation was mounted at the back door of a small van. During sampling, the van doors were open and the instrumentation faced the wind. Estimates of the numbers of Aitken particles were obtained using a General Electric small particle detector (type CN) whose calibration had been adjusted according to comparison with the readings of an absolute counter of the Scholz type (see Junge, 1966).

The size spectra of particles and droplets with radii from about 0.3 to 7 μ were obtained with a Royco particle counter, Model PC 200A (Royco Instruments, Menlo Park, Calif.). This instrument determines particle size from the amount of white light scattered by a particle at an angle of 90 degrees to the incident beam. It has some shortcomings which were noted in the earlier paper, as were the steps taken to minimize the effects of these shortcomings. The principal corrective measures were: to allow the short inlet tube to be exposed to the ambient air to minimize unnatural temperature fluctuations; and to make adjustments to the

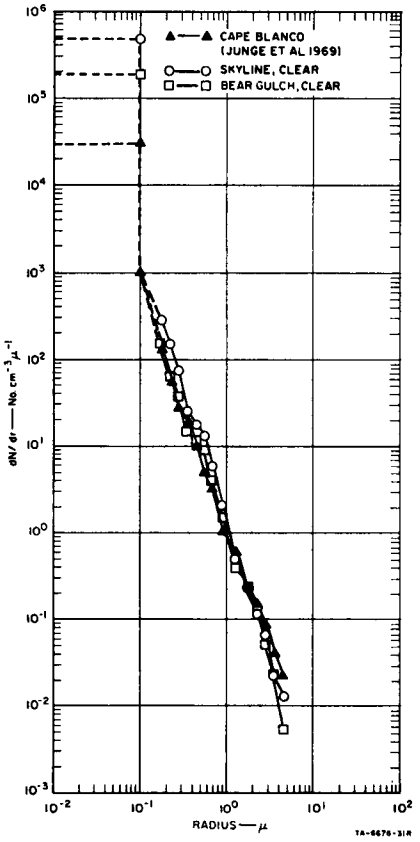

Fig. 1. Average particle size distributions on cloudless days.

manufacturer's calibration (using the findings of Hodkinson & Greenfield, 1965) to make it appropriate to the index of refraction for water.

The droplets larger than 8-μ radius were sized using a horizontal elutriator. This is a long, shallow tunnel through which the sampled air passes. The size distribution can be determined from the numbers of droplets that settle out at different distances from tunnel entrance. These numbers are determined from the impressions in magnesium oxide coated slides placed at different locations along the length of the tunnel. This method for determining size distributions is analogous to the methods that measure deposition rates at the bottom of a closed chamber (see, e.g., Cadle, 1955, pp. 204–8).

The air leaving the horizontal elutriator passed through two parallel Andersen cascade impactors (Andersen, 1958) and then through glass fiber filters. These cascade impactors classified, by size, those droplets and particles with radii less than about 5 μ into fractions for subsequent chemical analysis. The bounds on the size classes were taken to be defined by the radii of 50 percent collection efficiency (see Ludwig, 1968) as determined by Flesch et al. (1967). Corrections were made to compensate for the deposition in the elutriator.

Coulson & Cavanagh's (1960) titration cell was used for chloride analyses. This cell titrates the chloride ions with coulometrically generated silver ions. It does not distinguish among the Cl^-, Br^- and I^- ions. The analysis procedure for the sulfur compounds uses a stream of hydrogen and helium in a microcombustion furnace to reduce these compounds to hydrogen sulfide. The hydrogen sulfide is then quantitatively determined by iodimetric microcoulometric titration. The extraction procedures and the sulfur and chloride compound analysis are essentially the same as those described by Junge (1966). The nitrate analysis was done using the "modified" procedure of Kahn & Brezenski (1967).

Periodic measurements of wind and wet and dry bulb temperature were also made during the sample collecting and droplet measuring periods.

Results

As in the previous paper, size distributions have been presented in the dN/dr form (as opposed to the $dN/d\log_{10} r$ form). The unit used is number of particles per cubic centimeter per micron of radius interval. For averaging purposes, data were classified according to whether they were collected: (1) during clear conditions, (2) below a cloud, (3) at the lower edge of the cloud, (4) in a developed cloud, (5) during cloud formation, or (6) during cloud dissipation. These categories were assigned subjectively according to the observer's assessment of prevailing conditions. The settling tunnel data were often collected over extended periods during which conditions changed so that it was difficult to choose a category. In such cases, the category best typifying the collection period was subjectively assigned.

Average particle and droplet spectra

As in the earlier paper, the condensation nuclei (CN) counts were assumed to be made up of particles with radii between 0.01 and 0.1 μ, and the Royco size distributions were extrapolated to 0.1-μ radius. This treatment of the data was used by Junge (1966) and is intended only to approximate the gross features of the distribution. In the figures that follow, the dashed lines represent the extrapolated portions of the size distributions.

Size distribution observations were taken on several cloudless days at two locations in the hills south of San Francisco. The averages of these observations are shown in Fig. 1, along with the average size distribution obtained at Cape Blanco, Oregon, during an earlier program (Junge et al., 1969). About 16 hours total of Royco and CN sampling were compiled for the Skyline average; about 3 hours for the Bear Gulch; about 28 hours at Cape Blanco.

The average size distributions at all three locations are very nearly identical over the radius range from about 0.1 to 2 μ and are well represented by a power law (see Junge, 1963) with dN/dr proportional to $r^{-2.9}$. This suggests a consistency among aerosols whose trajectories bring them to the coast from over the North Pacific Ocean. The differences at the extremes of the distributions may be the result of local effects. For instance, the greater number of large particles at Cape Blanco may have come from nearby surf or sea spray sources, although this was undoubtedly minimized by the site's location 65 m above the sea surface.

The greater numbers of CN at the California sites may come from anthropogenic sources. Although there are very few sources between the sites and the sea, the area is not entirely free of them. Occasional high concentrations of CN indicated that there may also be some recirculation of Bay Area pollutants on the land-sea breeze cycle.

The averages from sampling at the Skyline site for a total of about four hours below the stratus and about five hours at its lower edge are shown in Fig. 2. The dashed portions of the graph at large radii indicate uncertainties in the settling tunnel results, generally because the concentrations were too low to be measured reliably. The concentrations of the smaller particles below the cloud and at its lower edge

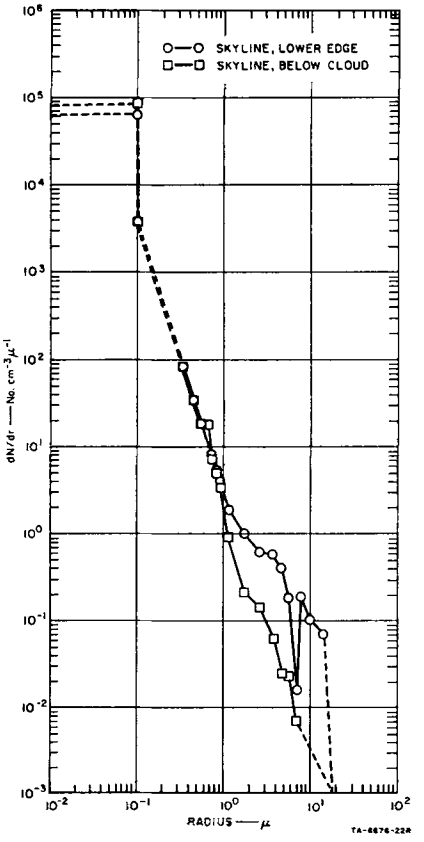

Fig. 2. Average size distributions below the stratus at Skyline.

are essentially identical. For radii between 0.3- and 1-μ, the concentration is approximately proportional to $r^{-3.1}$. This relationship also holds resonably well below the cloud for larger radii, but at the cloud's lower edge there is a tendency toward a secondary mode between about 2.5 and 3.5 μ.

An attempt was made to obtain data during periods of cloud formation and dissipation. Because most of the data collection took place during the morning hours, there is considerably more information available from periods of cloud dissipation than from periods of cloud formation. Curves representing the average size distributions during the formation and dissipation phases of cloud development are shown in Fig. 3 along with the average observed in developed clouds. About 2–½ hours of observations, mostly in the evening, are represented by the cloud formation average. About 7 hours

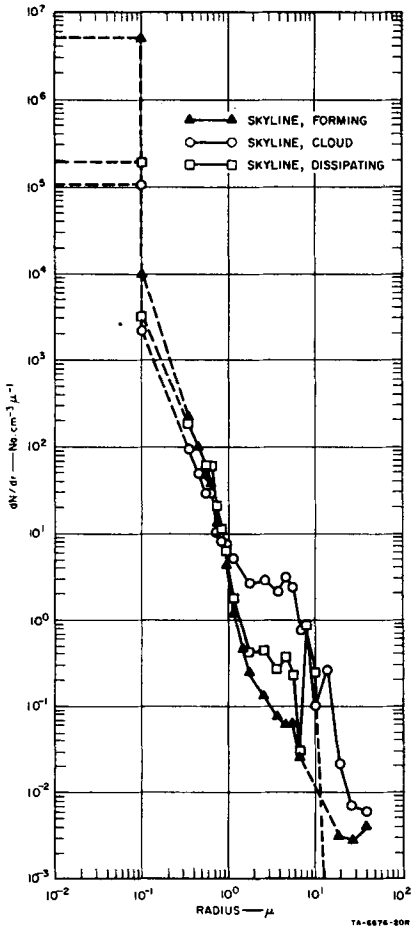

Fig. 3. Average particle size distributions at various stages of stratus development at Skyline.

of observations from mornings compose the cloud dissipation average. The average size distribution for developed clouds is based on about 36 hours of observation at the Skyline site.

The average size distribution for developed clouds shows a broad secondary mode around a radius of about 5 μ. The concentration at this secondary mode is about 3 droplets $cm^{-3}\mu^{-1}$. For sizes between 0.1 and 1 μ, the average particle concentrations in the developed cloud are approximately proportional to $r^{-2.6}$. The average size distribution observed during periods of cloud dissipation has a secondary mode at a radius of about 8 μ with a concentration of 0.9 $cm^{-3}\mu^{-1}$, about the same average concentration observed in the cloud at this radius. For radii between about 0.5 μ and 1.7 μ, the forming and dissipating cloud averages are very much alike and are strong functions of radius, being approximately proportional to $r^{-4.5}$. For sizes outside this range, the dependence is not so great. In general, there are more small particles during cloud formation than during dissipation and more of the larger droplets during dissipation than during formation.

It is not certain whether the very large numbers of CN observed during periods of cloud formation are representative. It appears, from the values observed under other conditions and at the Bear Gulch site, that they are not.

Examples of temporal changes in particle and droplet spectra

Although the average size distributions show the conditions for different stages of cloud development, they do not show the transition from one state of development to another. They also do not show the temporal variability and the nature of the changes that take place in the particle and droplet population of the developed cloud. To illustrate changes of this type, the Royco data were combined with CN and settling tunnel data to yield time sections of the particle and droplet spectra. In the following examples the isopleths have logarithmic intervals of frequency that correspond with the log–log graphs already presented. Times are given in local standard time (LST). The spectral frequencies at sizes larger than 7-μ radius are based on settling tunnel data, which represent periods ranging from about 20 min to about 2 hours, and therefore can only show the general nature of the size distribution changes. Wet and dry bulb temperatures measured to the nearest half-degree Fahrenheit are also shown in the figures. While inadequate for precise supersaturation measurements, they provide reasonable estimates of cooling rate or humidity at subsaturation conditions.

September 5, 1969, Morning. The observations on this day, as shown in Fig. 4, began with a very pronounced secondary mode at about 5- to 6-μ radius. At this mode, the concentration was greater than 10 droplets $cm^{-3}\mu^{-1}$, whereas the minimum at about 1.5 to 2 μ was less than 4 $cm^{-3}\mu^{-1}$. This droplet mode remained reason-

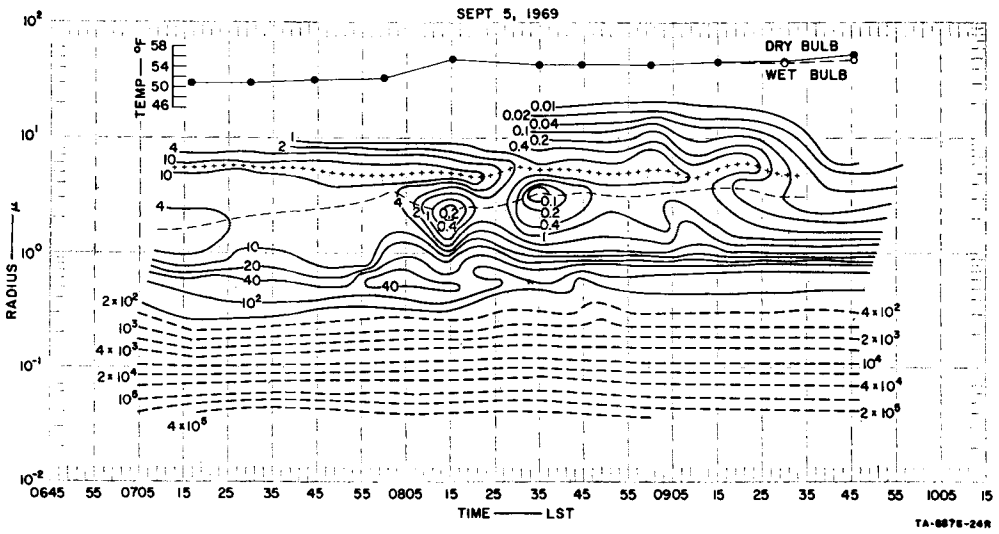

Fig. 4. Variations of the size distribution at Skyline, September 5, 1969, morning.

ably stable in magnitude and radius for about an hour, then the air temperature rose about 4°F in 15 min. This may have been caused by a temporary lowering of the inversion. The secondary mode during this period remained greater than 10 $cm^{-3}\mu^{-1}$, but there was a rapid loss of droplets in the 2- to 3-μ radius range that resulted in the formation of a deep minimum of less than 0.2 droplets $cm^{-3}\mu^{-1}$ at 2.3-μ radius. Changes in the size distribution at submicron sizes that caused the frequency distribution between about 0.3 and 0.8 μ to become nearly flat also occurred during this period.

Following the abrupt temperature rise, there was a slight drop in temperatures. At that time, the number of droplets in the 2- to 3-μ radius range increased again and the number from 4 to 7 μ decreased. The total effect was to reduce the sharpness of the 6-μ radius secondary mode. Around 0835, this mode became temporarily more pronounced again because of a decrease in the numbers of droplets with radii about 3 μ.

The air became unsaturated beginning at about 0915 LST. At this time, the decrease in the numbers of droplets with radii from 3 to 4 μ was greater than the decrease in the numbers of larger droplets so that there was a brief intensification of the secondary mode at a radius of about 6 μ and a concentration greater than 0.4 droplets $cm^{-3}\mu^{-1}$. However, the numbers of all particles with radii greater than about 1.5 μ soon decreased as the cloud dissipated.

September 11, 1969. This day is interesting because of the occurrence of a partial solar eclipse. About two-thirds of the sun's disc was obscured at the height of the eclipse. At the time sampling began (Fig. 5), the air was slightly unsaturated, and the secondary mode at about 5-μ radius was not very pronounced. The distribution was very nearly flat from about 0.8 μ to about 6 μ. The air became saturated, and a minimum of less than 1 droplet $cm^{-3}\mu^{-1}$ developed at about 2-μ radius, making the droplet mode more pronounced.

Some warming began around 0915, and the numbers of 5-μ and larger radius droplets began to decrease. At about the time of the beginning of the eclipse, the air became unsaturated, and by 0945 the air temperature reached a maximum. After this, cooling began, and the numbers of large particles increased again. Secondary modes developed, but they were not very pronounced. In fact, the distribution was nearly flat in the region from about 0.9 to 6 μ between 1030 and 1130. After this time the air began to warm again as the eclipse waned. The radius at which the secondary maximum occurred declined from about 4 μ to about 2.5 μ. Just before the end of the eclipse, the numbers of the droplets with radii greater than 1 μ quickly declined and the cloud disappeared.

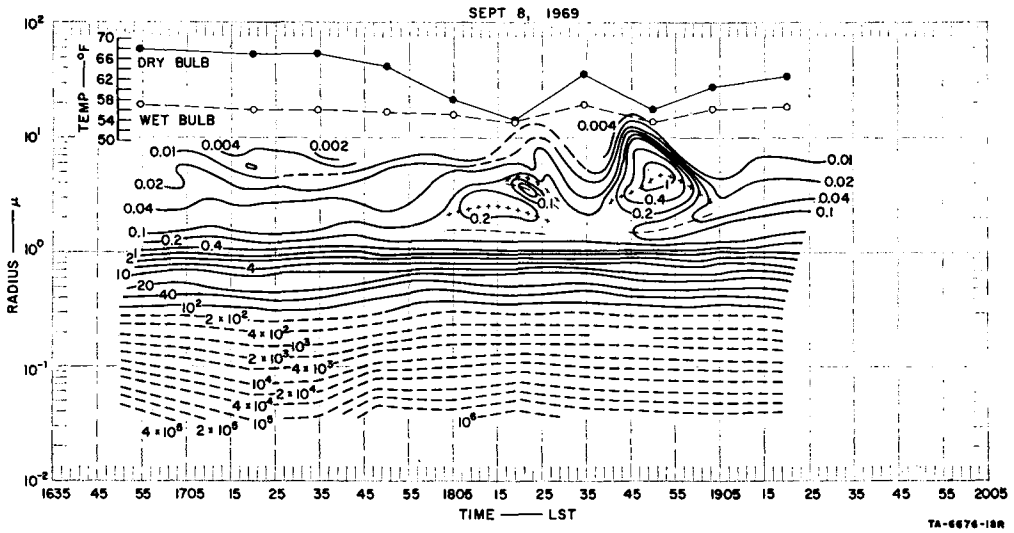

Fig. 7. Variations of the size distribution at Skyline, September 8, 1969, evening.

evening sampling period illustrated in Fig. 7, the stratus was well to the west of the Skyline site, having penetrated only a short distance inland from the coast. The air was relatively dry, indicating that the site was in the inversion layer above the marine air. The stratus gradually penetrated farther and farther up the canyon and through the col to the north of the site. During this period the inversion appears to have lifted, and the temperature dropped as the marine air began to envelop the site. With the influx of marine air, the first cloud was observed at about 1815.

The onset of the fog produced almost no change in the size distribution for particles with radii between about 0.1 to 1 μ, but as is to be expected, there were increases in the numbers of larger particles. The greatest increases occurred between about 1 and 7 μ.

As the inversion lowered again, the cloud quickly dissipated and droplets larger than about 3-μ radius disappeared. One more incursion of marine air and cloud took place, accompanied by the development of a sharp but short-lived secondary mode at a radius of about 5 μ. As the cloud withdrew, the size and the numbers of particles constituting this secondary mode declined. The stratus withdrew to lower levels, generally toward the west, although some still penetrated the col to the north.

Chemical constituents

The samples collected by the Andersen cascade impactors yielded data on the size distributions of different chemical constituents. Three different constituents will be discussed: sulfur-containing compounds, chlorides, and nitrates. We analyzed for sulfur and chloride compounds in samples collected during both summers, 1968 and 1969. The nitrate compounds were studied only during the summer of 1969.

The average mass distributions for these constituents are shown in Fig. 8. The sulfur and chloride results for the six stages of the Andersen sampler, i.e., sizes greater than 0.35-μ radius, represent the size distribution found in 165 to 178 m^3 of air. The differences in sampled volume are the result of lost samples (see Ludwig & Robinson, 1970). The nitrate results shown in Fig. 8 are based on the sampling of about 85 m^3 of air during morning and evening hours of 1969.

As found from the earlier studies (Ludwig & Robinson, 1970), the distribution of sulfate with particle size more or less follows the droplet size distribution, with greater amounts of sulfate being found at the smaller sizes, minimum amounts around 2-μ radius and a secondary maximum at about 3 μ. The nitrate materials are distributed similarly. The activities of the

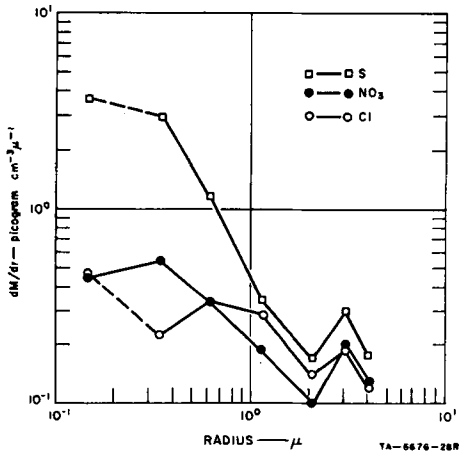

Fig. 8. Average mass distributions of three chemical constituents.

nitrates and the sulfates as droplet nuclei appear to be size dependent; the smaller particles are inactive and grow little while the larger particles grow to larger droplets. A minimum is then left in the spectrum between the growing droplets and the inactivated particles. The average distribution of chloride material with particle size is similar to those of the other two constituents, but the variations are not so pronounced.

Discussion

In the paper describing the earlier results (Ludwig & Robinson, 1970), the average spectra were compared with those calculated from theory by Neiburger & Chien (1960). The comments concerning that comparison are still valid. The calculations and the theory are similar in that both show modes in the Aitken nuclei size range and at radii of a few microns. They differ principally in spectral concentration at the larger radius mode, with the calculated values several tens of times larger than the average observed values. The calculations also predict a very sharp mode at these larger droplet sizes, while the average of the observations shows a very broad mode.

Some of these differences can be attributed to the averaging process, which tends to smooth the sharp features of the individual size distributions. The time-section examples given above show some fairly sharp modes at the large droplet sizes. The individual spectral frequencies often exceed 10 droplets $cm^{-3}\mu^{-1}$, which is within an order of magnitude of the calculated values.

Other reasons for the differences between calculations and observations center on the difference between the assumptions used in the calculations and the actual conditions in the atmosphere. In the earlier paper, we pointed out that the cooling rate used by Neiburger and Chien is much more rapid than was generally observed during our sampling and this could contribute to the differences. Other possible sources of the differences include mixing with air outside the cloud (Mason, 1960), inhomogeneous distribution of cloud condensation nuclei concentration within the cloud, or mixed composition of cloud condensation nuclei (Kornfeld, 1970).

The time sections illustrate another difference between observation and the calculated results. The radii at which the secondary modes are found in the time sections are not observed to increase regularly with time as suggested by the calculations. This is probably due, in part, to the fact that the fog develops at the site through a combination of cooling and advection. Thus, fog with a reasonably mature droplet size distribution is brought to the site complete with a developed secondary mode. Cooling does play some part, because there is lifting as the air passes over the hill and because there is cooling of the air and underlying surfaces by radiation. However, from the nature of the development of the size distribution and from subjective observation at the site, advection appears to be a dominant factor in the onset of fog at the Skyline location. The fact that modes were very seldom observed at very large sizes, even in mature morning fogs, suggests that the droplet mode does not continue to migrate toward larger sizes with time.

A number of observational studies were cited in our earlier paper, and these tended to support the conclusion that the droplet mode does not continue to move to larger and larger sizes, For those studies where modes were found at larger droplet sizes, the radii at which they occurred were usually less than 12 or 13 μ. Similar results have been found in more recently reported studies (Kocmond et al., 1970; Kunkel, 1970).

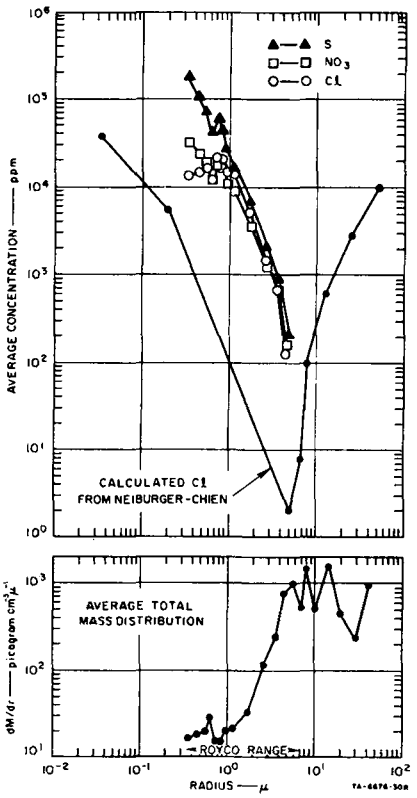

Fig. 9. Total mass and concentration distributions with size.

Inasmuch as chemical data are available, it is possible to compare the concentrations of chloride material in the various size droplets as determined from the Neiburger-Chien calculations with those concentrations of the constituents observed in the stratus. Fig. 9 shows the average distribution of droplet mass based on Royco and settling tunnel data. This total distribution assumes the particles and droplets to be of unit density and spherical and was based on the average number distribution for all the different types of cloud conditions, not including the clear days. Some data are also included from the "Bear Gulch" site (see Ludwig & Robinson, 1970), a few kilometers farther to the south and at a somewhat higher elevation. The chemical data were also collected for all types of cloud conditions and at the two locations. The upper part of Fig. 9 combines the average mass distribution with the data from Fig. 8 to determine the dissolved concentrations of three different ions as functions of droplet size. The slopes of these curves are greater than -3, so that larger amounts of the materials are present in the nuclei of the larger droplets. Also shown in the figure is the concentration curve based on the Neiburger–Chien (1960) calculations. This calculated curve used the droplet volumes at 3 000 sec after cooling began and the volume of the corresponding nuclei. This volumetric concentration was adjusted for density and atomic weight of chloride to convert to the concentration curve shown. The droplet size at 3 000 sec was chosen because that spectrum is more like the observed sizes than are the 6 000-sec values.

The calculated concentration is about two orders of magnitude less than the observed from about 0.7- to 5-μ radius. This implies that the nuclei required in nature to produce given droplet sizes are larger than those calculated by Neiburger & Chien. Considering the slower cooling rates during the observations and the larger numbers of small nuclei competing for the available condensing water vapor, it isn't too surprising that larger nuclei might be required. The available data are not sufficient to confirm or deny the Neiburger–Chien prediction of an increae in the concentration of nucleus material for the larger droplet sizes.

The average ratios of sulfur to chloride and of nitrate to chloride are found in Fig. 10. Sea water composition values given by Sverdrup et al. (1942) indicate that the sea water ratio of nitrate to chloride should be about 10^{-4} and the sulfur to chloride ratio about 4.7×10^{-2}. Both values are well below those shown in Fig. 10. It appears that sea spray is not the dominant aerosol in the area studied, although this area is only a few kilometers downwind of the ocean and virtually all sampling was done within the marine air mass.

Holzworth (1959), Lodge et al. (1960), and Junge et al. (1969) have all studied the composition of aerosols in maritime air masses from the north Pacific. Holzworth's data were collected at two California coastal sites and two islands off the California coast. The Lodge data came from weather ship November (latitude 30° N, longitude 140° W). Junge's observations were made at Cape Blanco, Oregon. The sulfur aerosol concentrations found at all these sites were comparable to those concentrations collected in the Andersen

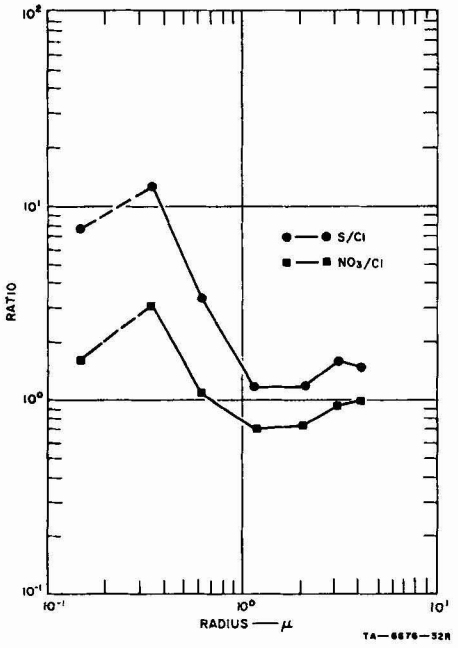

Fig. 10. Sulfur-to-chloride and nitrate-to-chloride ratios as functions of size.

samples of this program, about 3 μg m^{-3}. Junge's two Cape Blanco measurements averaged about 1 μg m^{-3}, and the median of Lodge's observations was about the same. Holzworth's observations of marine aerosols averaged about 3 to 6 μg m^{-3} of sulfur (as sulfate).

The average nitrate aerosol concentration observed on this program was about 1 μg m^{-3}. The median of the observations at weather ship November was slightly less than 0.2 micrograms per cubic meter. The average concentration that Holzworth found in air of maritime origin at his California coastal sites ranged from 1 to 3 μg m^{-3}.

The Andersen sampler collections of this program showed an average chloride concentration of about 1 μg m^{-3}. Junge et al. (1969) measured about 25 μg m^{-3} of chloride at Cape Blanco, and Holzworth's (1959) average observations in maritime air masses ranged from about 5 to 120 μgm m^{-3}. The weather ship November data (Lodge et al., 1960) had a median chloride concentration of about 2 μg m^{-3}. Surf effects may account for the higher chloride concentrations observed by Holzworth and by Junge. For example, Holzworth states that the low chloride values that were observed at one of his sampling locations (San Nicholas Island) "may be attributed to the elevation of the sampling site, 500 ft (150 m), well removed from the surf spray". Although the data of Junge et al. were collected at an elevation of about 65 m, there is some evidence of surf effects; e.g., more than 90% of the collected chloride was contained in particles with radii larger than 1 μ.

All the above data suggest that the high chloride concentrations are generated near the shore and that there is a subsequent loss of chloride in the atmospheric aerosol. The data also indicate that there are important sources, besides sea salt, of sulfur and nitrate compounds, even in areas uncontaminated by industrial sources. Sulfur-to-chloride and nitrate-to-chloride ratios greater than those found in sea water are the rule rather than the exception.

It was noted in our earlier paper that Robbins et al. (1959) had proposed an atmospheric chemical reaction that could account for a chloride deficit. They suggested that nitric acid vapor was formed by the hydrolysis of nitrogen dioxide. Nitric acid vapor is quickly absorbed by the solution droplets. It reacts with the sodium chloride to release hydrogen chloride vapor. The high nitrate content found in the droplets tends to support this theory, but the data might also support an hypothesized process whereby sulfate replaced chloride.

The laboratory work of Robbins et al. indicates that the process of chloride removal by nitrate formation is more efficient in the more dilute droplets. This suggested (Ludwig & Robinson, 1970) that the replacement of chloride by nitrate should be size dependent. If this were true, then the nitrate-to-chloride ratio might be expected to increase with decreasing chloride concentration in the droplets. Comparison of the observed and calculated droplet concentrations of chloride shown in Fig. 9 with the nitrate-to-chloride ratio shown in Fig. 10 does not support the hypothesis of a strong inverse relationship between the concentrations of chloride and nitrate materials in the droplets.

The results of these studies have provided good descriptions of the droplet and aerosol size distributions in the marine air mass stratus that are found in the hills south of San Francisco, and of the changes in these

distributions during the onset and dissipation of the clouds. Substantial information has also been gained about the chemical nature of the droplets and aerosols, particularly the chloride, nitrate, and sulfur compounds. Some questions have been raised by the research, particularly regarding the fate of the chlorides and the origins of the other nuclei constituents studied. This paper has suggested a possible mechanism that might account for relatively low chloride concentrations and we intend to investigate this and reactions involving sulfur compounds further through laboratory studies.

The observed size distributions differ somewhat from those found by numerical modeling of the condensation processes. This suggests that the hypothesized conditions used in the modeling have not been as realistic as they might have been. Possible significant changes include cooling rates less than 6°C hour^{-1} as used by Neiburger & Chien; irregular, rather than constant, cooling rates; and a nuclei population of mixed composition rather than wholly sodium chloride. These factors could probably be incorporated reasonably easily in existing models. Another potentially important factor, not so easily included in current models, is the effect of entrainment of unsaturated air into the cloud and the loss of droplet-laden air from the cloud. We hope the future research with numerical modeling will be able to tell us something of the importance of the above factors to the size distribution that develops in stratus clouds.

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REFERENCES

Andersen, A. A. 1958. A new sampler for the collection, sizing, and enumeration of airborne particles. *J. Bacteriol. 76*, 471–484.

Cadle, R. D. 1955. *Particle size determination*, 303 pp. Interscience Publishers, Inc., New York.

Coulson, D. M. & Cavanagh, L. A. 1960. Automatic chloride analyzer. *Anal. Chem. 32*, 1245–1247.

Flesch, J. P., Norris, C. H. & Nugent, A. E., Jr. 1967. Calibrating particulate air samplers with monodisperse aerosols: Application to the Andersen cascade impactor. *J. Am. Industr. Hyg. Assoc. 28*, 507–516.

Hodkinson, J. R. & Greenfield, J. R. 1965. Response calculations for light-scattering aerosol counters and photometers. *Appl. Optics 4*, 1463–1471.

Holzworth, G. C. 1959. Atmospheric contaminants at remote California coastal sites. *J. Meteorol. 16*, 68–79.

Junge, C. E. 1963. *Air chemistry and radioactivity*, 382 pp. Academic Press, New York.

Junge, C. E. 1966. Studies of the chemistry of unpolluted atmospheres. Final Report, U.S. Wea. Bur. Contract No. Cwb-11151, Menlo Park, Calif., Stanford Research Institute, 98 pp. Available Fed. Clearinghouse Sci. and Tech. Infor. No. AD 646 465.

Junge, C. E., Robinson, E. & Ludwig, F. L. 1969. A study of aerosols in Pacific air masses. *J. Appl. Meteorol. 8*, 340–347.

Kahn, L. & Brezenski, F. 1967. Determination of nitrate in estuarine waters. Comparison of a hydrazine reduction and a brucine procedure and modification of a brucine procedure. *Environ. Sci. and Tech. 1*, 488–491.

Kocmond, W. C., Pilie, R. J., Eadie, W. J. & Mack, E. J. 1970. Recent fog modification research at Cornell Aeronautical Lab. *Proc. Second Nat. Conf. on Wea. Modif.*, Santa Barbara, Calif. April 6–9, 1970, pp. 112–116.

Kornfeld, P. 1970. Numerical solution for condensation of atmospheric water vapor on soluble and insoluble nuclei. *J. Atmos. Sci. 27*, 256–264.

Kunkel, B. 1970. Fog drop-size distributions measured with a laser hologram camera. *Proc. Conf. on Cloud Physics*, Ft. Collins, Colo., Aug. 24–27, 1970, pp. 95–96.

Lodge, J. P., MacDonald, A. J. & Vikman, E. 1960. A study of the composition of marine atmospheres. *Tellus 12*, 184–187.

Ludwig, F. L. 1968. Behavior of a numerical analog to a cascade impactor. *Environ. Sci. and Tech. 2*, 94–105.

Ludwig, F. L. & Robinson, E. 1969. Condensation nuclei and aerosol populations related to fog formation. Final Report, Army Research Office-Durham Contract No. DAHC04-67 C-0059, Menlo Park, Calif., Stanford Research Institute, 98 pp. Available Fed. Clearinghouse Sci. and Tech. Infor. No. AD 700137.

Ludwig, F. L. & Robinson, E. 1970. Observations of aerosols and droplets in California stratus. *Tellus 22*, 94–105.

Mason, B. J. 1960. The evolution of droplet spectra in stratus cloud. *J. Meteorol. 17*, 459–462.

Neiburger, M. & Chien, C. W. 1960. Computations of the growth of cloud drops by condensation using an electronic digital computer. *Geophys. Monograph No. 5*, 191–209.

Robbins, R. C., Cadle, R. D. & Eckhardt, D. L. 1959. The conversion of sodium chloride to hydrogen chloride in the atmosphere. *J. Meteorol. 16*, 53–56.

Sverdrup, H. V., Johnson, M. W. & Fleming, R. H. 1942. *The Oceans*, 1087 pp. Prentice-Hall, Inc., New York.

НАБЛЮДЕНИЯ АЭРОЗОЛЯ И КАПЕЛЬ В ПЕРИСТЫХ ОБЛАКАХ НАД КАЛИФОРНИЕЙ (II)

Летом 1968 и 1969 гг. на холмах к югу от Сан-Франциско наблюдались распределения по размерам для аэрозолей и капель. В число приборов входили счетчик частиц по рассеянию света Ройко, счетчик ядер конденсации и заборная трубка. Осредненные распределения по размерам проявляют основные пики в субмикронной области для ясных дней, в периоды образования облаков и их рассеяния и в развитых облаках. Широкий вторичный пик порядка 3 капель на $см^{-3}$ μ^{-1} был найден для радиуса около 5 μ в облаках. Несколько более резкий пик порядка 0,9 $см^{-3}$ μ^{-1} при радиусе 8 μ был найден в осредненных распределениях по размерам для рассеивающихся облаков. В периоды развития облаков этот вторичный пик выражен слабо. Дается временное развитие спектров частиц и капель, что указывает на значительные временные вариации спектров. Анализ временного развития не подтверждает общей тенденции увеличения размеров капель со временем, как это предсказывалось некоторыми численными расчетами конденсационных процессов. Представлены результаты анализов содержания хлоридов, серы и нитратов в аэрозолях и капель. Количество серных компонентов и нитратов по отношению к хлоридам слишком велико, чтобы основным источником этих веществ была бы морская соль, если не считать, что хлориды были удалены путем каких-то химических процессов.