Chemistry of Aerosols, Cloud Droplets, and Rain in the Puerto Rican Marine Atmosphere

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The chemical changes occurring during the transformation of aerosols to cloud droplets and rain were observed in orographic clouds on the eastern end of the island of Puerto Rico. Comparisons of elemental ratios in particles and rain and of elemental particle size distributions in and out of the clouds demonstrated scavenging efficiencies of >100% for the elements I, Br, and Cl relative to Na and of <25% for the elements Al, Mn, and V relative to Na. These different scavenging efficiencies are consistent with predictions based on existing cloud physics theory and are a reflection of the geochemical properties of the aerosol consisting partly of a hygroscopic sea salt component containing I, Br, Cl, and Na and partly of a soil dust component containing Al, Mn, and possibly V.

This paper uses chemical measurements to document the processes through which aerosols are transformed into or incorporated by cloud droplets and rain during natural cloud formation.

Several field experiments directly tracing growth of aerosols to cloud droplets have been carried out, generally through microscopic studies of cloud droplet particle size distributions [*Warner*, 1969; *Ludwig and Robinson*, 1970, 1971; *Garland*, 1971]; however, little work has been done on relating the actual chemical composition of cloud droplets and rain to the chemical composition of nucleating aerosols through simultaneous collection of all three in clouds.

The measurements reported here are designed to test the reliability of existing theories concerning the behavior of particles during cloud formation. Comparisons of chemical composition of simultaneously collected aerosols, cloud droplets, and rain and observations of differences in the particle size distributions of elements in and out of clouds are used. Considerable knowledge of the chemistry of marine aerosols and rain attained through previous studies [i.e., *Woodcock and Blanchard*, 1955; *Duce et al.*, 1965, 1967] provides a firm basis

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for these experiments, particularly for the halogens I, Br, and Cl. Recently developed methods of sensitive nondestructive neutron activation analysis now allow quantitative measurement of many formerly undetectable elements and do not require any radiochemical steps or elemental separations [*Dams et al.*, 1970].

APPLICATION OF CLOUD PHYSICS THEORY

Warm atmospheric clouds are formed by the condensation of water vapor on aerosols. Variables important in considering individual particles as potential cloud nuclei are solubility and size.

In this work both hygroscopic and nonhygroscopic particles over the diameter range 0.01 to >10 μ were studied. The major theoretical factors of importance to the investigation were that (1) uniform cloud droplet size spectra result at cloud base during cloud formation regardless of the size spectrum of nucleated aerosols [*Mordy*, 1959; *Neiburger and Chien*, 1960; *Warner*, 1969] and (2) cloud droplets should not appreciably scavenge unnucleated aerosols [*Greenfield*, 1957; *McDonald*, 1964; *Shafrir and Neiburger*, 1963; *Bartlett*, 1970; *Hocking and Jonas*, 1970].

Condensation theories predicting the potential of both hygroscopic and nonhygroscopic aerosols as nuclei have been developed. *Mordy* [1959], following the earlier computations of *Howell* [1949], modeled the condensation growth of

populations of hygroscopic sea salt nuclei. *McDonald* [1964] gave a detailed theoretical discussion on condensation growth of insoluble (nonhygroscopic) particles. The predicted cutoff diameter for nucleation potential of hygroscopic particles occurred in the particle diameter range of 0.1 to approximately 1.0 μ, whereas only nonhygroscopic silicate dust particles larger than several microns in diameter were expected to act as nuclei.

Field observations of cloud droplet populations by *Rosinski* [1967], *Warner* [1969], and *Ludwig and Robinson* [1970, 1971] show some agreement with nucleation predictions for both types of particles, and cloud droplet number concentrations have been shown by *Twomey and Warner* [1967] and *Warner* [1969] to be closely related to number concentrations of nuclei below cloud base. *Warner* [1969] has further demonstrated the growth of droplets toward a uniform diameter in cumulus clouds.

The narrow size distribution range of small cloud droplets produced by condensation growth is broadened to include larger droplets by collisions and coalescence of existing droplets [*Berry*, 1967] as well as by further condensation. The simultaneous effects of collisions between cloud droplets and their growth by condensation leading to raindrops have been considered in detail by many authors including *Warshaw* [1967], *Berry* [1967], *Scott* [1968], *Kornfeld et al.* [1968], and *Kovetz and Olund* [1969]. Their work has produced numerical growth equations based on the probability of occurrence of any potential coalescence or condensation growth in real clouds.

Once they are grown, cloud droplets should not scavenge unnucleated large and giant aerosols with any appreciable efficiency in terms of transfer of chemical mass [*Greenfield*, 1957; *McDonald*, 1964]. Aerodynamic capture efficiencies are shown by *Shafrir and Neiburger* [1963], *Davis* [1966], *Warshaw* [1968], *Bartlett* [1970], and *Hocking and Jonas* [1970] to be inappreciable for large and giant aerosols, and collection due to Brownian motion diffusion is unimportant for particles with diameters greater than about 0.02 μ [*Byers*, 1965].

This combination of experimental and theoretical evidence suggests that, although the absolute concentration of individual elements in cloud droplets and rain depends on condensa-

tion, coalescence, and evaporation processes, the ratio of elements should be the same as that in scavenged sea salt and dust particles if there was no absorption of gaseous phases of the elements or selective elemental volatilization.

EXPERIMENTAL PROCEDURE

Study area and sampling sites. The eastern end of the island of Puerto Rico was chosen for this study owing to the relatively reproducible meteorological conditions encountered there [*Odum et al.*, 1970] and the possibilities for obtaining large aerosol and cloud droplet samples requiring long sampling times both in and out of clouds. The relatively reproducible aerosol population sampled there [*Andren*, 1971] was carried inland by the easterly trade winds before encountering a ridge of mountains including the easternmost Pico del Este. Typical orographic cumuli enveloped the top of this peak during approximately 75% of the entire expedition during late November 1971.

Four sampling stations as shown in Figure 1 were located on the coastline at a Navy base (NC), just below cloud base at a boy scout camp (BS), on top of Pico del Este in a cloud forest (CF), and on the leeward side of the mountain ridge in the rain forest (RF). The samplers at stations other than RF were held by ring stands 1 meter above the roofs of one- or two-story inactive buildings. The RF sampler was located on a 22-meter tower above the rain forest tree canopy.

Owing to complexities in the topography and micrometeorology of this area it was not pos-
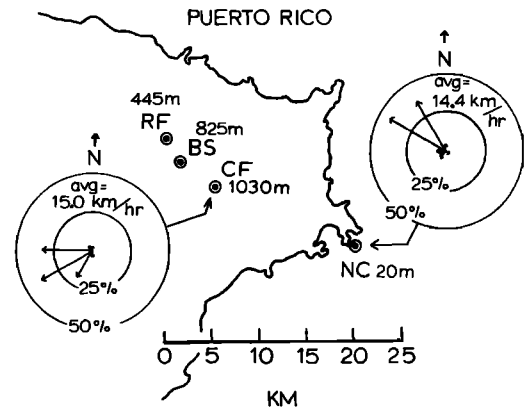

Fig. 1. Station locations and averaged wind data.

sible to determine the ages of the particles reaching the various stations, although those from the NC station were undoubtedly the freshest in terms of the sea salt component.

Sampling methods and materials. The instruments used for sample collection were (1) Andersen cascade impactors in combination with 47-mm afterfilters, (2) a gravitational settling tunnel with an Andersen impactor and afterfilter, and (3) nalgene funnel rain collectors.

To study particle growth during cloud formation at station CF, it was necessary to simultaneously separate and collect aerosols and cloud droplets at cloud base. This process was accomplished through the use of a gravitational settling tunnel mounted directly on an Andersen impactor with afterfilter. The gravitational settling tunnel followed the basic design of *Ludwig and Robinson* [1970]; however, it was modified to be used with a single Andersen impactor at a flow rate of 1.7 m³/hr. Cloud droplet particles settled out on 0.0025-cm-thick polyethylene covering a glass plate. The glass plate was mounted on a removable tray in order to facilitate handling of the samples.

Collection of rain droplets by the settling tunnel was avoided by mounting a plexiglass plate on two bolts 7 cm above the tunnel intake.

The settling tunnel removed appreciable amounts of particles that normally would have landed on the 1 and 2 stages of a free Andersen impactor well in agreement with Stokes settling theory. Uncorrected and corrected results reported here (Figure 2) were based on simultaneous tests of impactors with and without this settling tunnel. Experimental details are presented by *Martens* [1972].

Collection of aerosols by using Andersen impactors has been discussed in detail by *Andersen* [1966], *Flesch et al.* [1967], *Gillette and Winchester* [1972], and *Hu* [1971]. Thin polyethylene disks placed on the Andersen sampler plates were used as impaction surfaces in all our experiments. A detailed discussion on the use of such materials has been presented by *Dams et al.* [1972]. The 47-mm afterfilters were added in order to sample a full range of particle sizes. Whatman 41 cellulose fiber paper was used in the afterfilter because it allows a fast flow rate, has good retention for submicron particles [*Lindeken et al.*, 1963], and is generally clean. All sampling was done at a flow rate of 1.7

m³/hr with vacuum pumps capable of self-adjustment to maintain constant flow (Radeco, Inc., Pleasant Hill, California). Twenty-five cm diameter plexiglass disks incorporated as part of the holders protected Andersen impactors at stations NC, BS, and RF from sporadic showers. Simultaneous tests of impactors with and without these holders showed no effects on aerosol collection.

During sampling, station CF was continuously attended, and the in-cloud nalgene funnel rain samplers were only uncovered during periods of rainfall. Andersen impactors and rain samplers at stations NC, BS, and RF were inspected and serviced on an almost daily basis.

Backup meteorological data were provided by portable recording hygrothermographs, sling psychrometers, and wind speed and direction gages.

Analytical methods. Aerosol, cloud droplet, and rain samples were simultaneously analyzed for I, Br, Cl, Na, Al, Mn, and V by nondestructive neutron activation analysis (NAA) with the 3-Mw reactor of the Lawrence Livermore Laboratory (Livermore, California). A high-resolution lithium-drifted germanium detector was used to count the samples. Details of the counting sequence used have been given by *Rahn et al.* [1971]. Rain samples were analyzed for Na by both NAA and atomic absorption (AA). Following analysis by AA the rain samples were concentrated by low-temperature evaporation and spotted on Whatman 41 paper for NAA. Comparisons of results can be seen in Table 2.

Details on corrections for polyethylene and Whatman 41 blanks are presented by *Martens* [1972].

RESULTS

Listing of results. A listing of the station samples, sampling periods, total volume sampled, humidity data, and total particulate masses of each of the elements is presented in Table 1. A listing of the nanograms per cubic meter of each element versus the Andersen impactor stage in each Puerto Rican sampling run has been given by *Martens* [1972]. Wind rose data from stations NC and CF are presented in Figure 1. Summarized information on rain samples is presented in Table 2. The letters A and R in the sample numbers refer to aerosol or rain

sample collection at the station, whereas the numerals 1 and 2 indicate two separate and nearly simultaneous sets of collections at the stations. Aerosol and rain samples with the same station prefix and numeral were sampled during the same time period.

Elemental particle size distributions. Figures 2 and 3 are plots of mass per cubic meter of each of the elements studied versus the Andersen impactor stage at the four sampling sta-

tions during the second nearly simultaneous collection (2). Details from the other run (1), as well as complete tabulated data for all Andersen runs, have been presented by *Martens* [1972]. The 50% cutoff diameters based on calibration of the Andersen impactor with unit density latex spheres and often used to describe the size of particles sampled by each stage [*Flesch et al.*, 1967] are included in Figure 3.

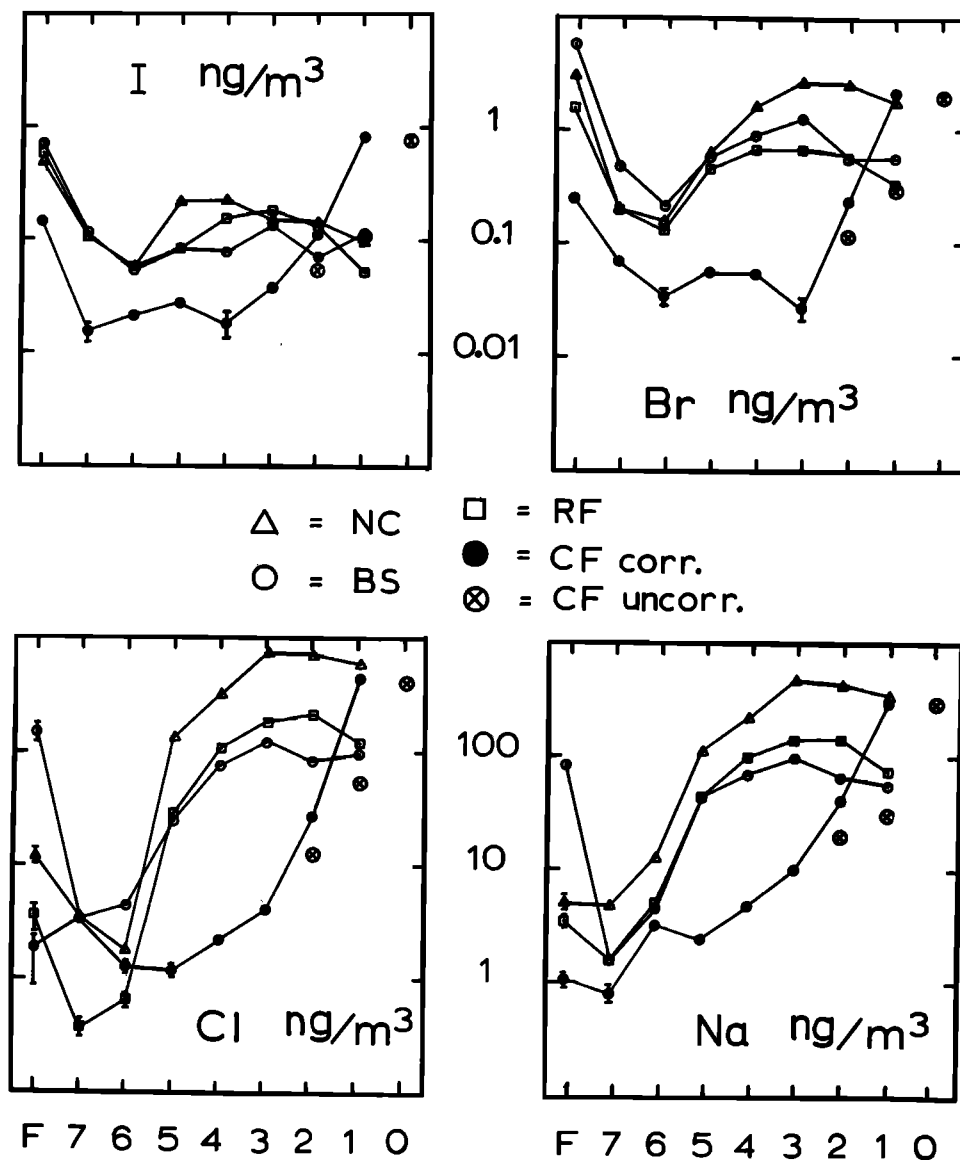The masses and elemental particle size dis-

Fig. 2. Particle size distributions of I, Br, Cl, and Na from simultaneous collection 2. Analytical uncertainties are approximately size of symbols.

TABLE 1. Puerto Rico Aerosol Sampling Summary

Aerosol Sample	Sampling Period, 1971	Sampling Volume, m³	Average Humidity, %	Total Particulate Elemental Concentrations, ng/m³						
				I 20%*	Br 15%*	Cl 10%*	Na 10%*	Al 20%*	Mn 20%*	V 20%*
NCA1	1200 Nov. 23 to 1300 Nov. 25	83	69	1.4	6.7	2500	1500	58	0.55	0.62
NCA2	1345 Nov. 25 to 1515 Nov. 28	125	70	1.5	12	2440	1500	24	0.21	1.7
BSA2	0950 Nov. 27 to 1400 Nov. 30†	74	84	1.4	10	550	400	190	12	2.2
RFA1	1015 Nov. 23 to 0710 Nov. 26	117	86	1.5	4.2	1890	1170	19	0.70	1.1
RFA2	0745 Nov. 26 to 1100 Nov. 30	169	80	1.4	4.5	625	475	22	0.83	2.7
CFA1	1435 Nov. 22 to 0900 Nov. 25§	64	100	1.9	10	1600	1000	270	6.6	2.4
CFA2	1845 Nov. 25 to 0830 Nov. 30§	125	100	1.2	2.8	465	350	30	1.6	1.0

*Approximate analytical uncertainty.
†Sampler shut down during cloudy periods.
§Sampler shut down during noncloudy periods.

tributions of I, Br, Cl, and Na are consistent with a sea salt aerosol source. The Al, Mn, and V are assumed to be from crustal material owing to their close agreement with crustal ratios [Martens, 1972] and low concentrations in sea water relative to Na. The similar behavior of these three elements during cloud formation and rainout further justifies this assumption. The Na contributions from crustal material based on average crustal ratios appear to be a few percent or less of the total Na except in the case of aerosol samples BSA2 and possibly CFA1. An interesting decrease in the Cl/Na ratio of the aerosols [Martens, 1972] appears to be due to Cl loss from particles rather than a soil addition of Na.

In-cloud shifts of distributions. Careful visual comparisons of the elemental particle size distributions at the coast, on the mountainside below cloud base, and over the rain forest with the in-cloud distributions on top of Pico del Este make possible certain generalizations about

TABLE 2. Puerto Rico Rain Sampling Summary

Rain Sample	Total Rain, mm	Elemental Rain Concentrations, µg/l							
		I 10%§	Br 10%§	Cl 5%§	Na* 5%§	Na† 5%§	Al 15%§	Mn 15%§	V 15%§
NCR2	4.3	1.7	25	8650	3160	3220	3.2		0.22
RFR1	19.6	2.9	13	3780	2210	2450	3.3		0.19
RFR2-1¶	26.7	1.4	14	360	245	250	11	0.67	6.4
RFR2-2¶	8.4	4.1	7.7	1420	655	600	3.5	1.4	0.23
CFR1	22.4	3.4	11	2550	1140	1070	1.4	1.7	0.30
CFR2	14.5	6.2	12	2300	1100	1110	4.6	0.80	0.66
CFSR2-1#	0.3	5.9	16	3750	1700	1680	9.5	1.3	0.49
CFSR2-2#	0.3	5.1	13	2550	1190	1170	6.6	0.72	0.66
CFSR2-3#	0.3	6.1	13	2350	1040	1000	5.7	0.82	0.89
CFSR2-4#	0.3	5.1	12	2170	995	1100	6.3	0.97	0.71
CFSR2-5#	0.3	4.1	13	2320	1050	1020	7.6	0.64	0.64
CFSR2-6#	0.1	4.8	13	2310	1100	1220	6.7	0.92	1.3

*Na by neutron activation.
†Na by atomic absorption spectroscopy.
§Approximate analytical uncertainty.
¶RFR2-1 and RFR2-2 collected sequentially during RFA2 aerosol collection.
#CFSR2-1 through CFSR2-6 collected sequentially from a single shower during. CFA2 aerosol collection.
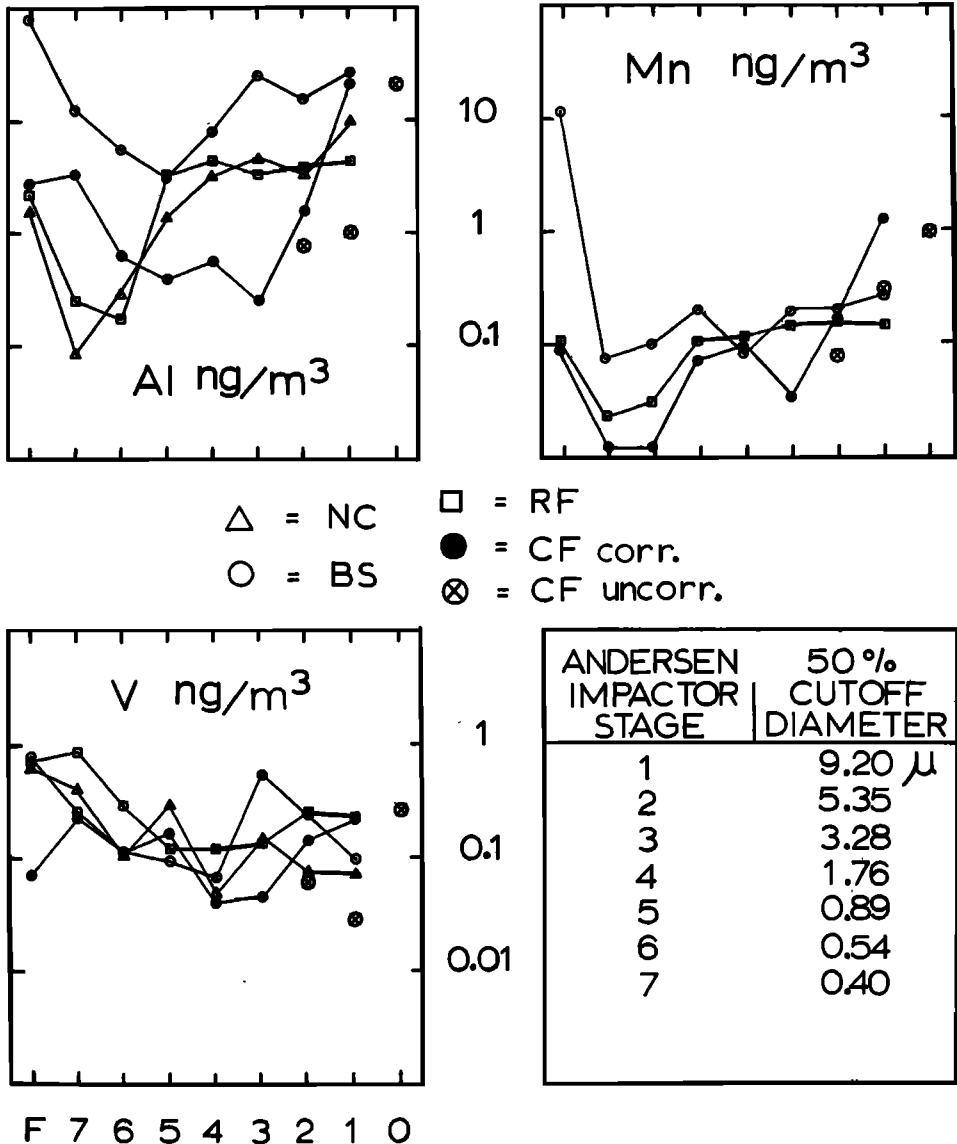
Fig. 3. Particle size distributions of Al, Mn, and V from simultaneous collection 2 and Andersen impactor 50% cutoff diameters. Analytical uncertainties are approximately size of symbols.

the behavior of each element during cloud formation.

Size distributions of I, Br, Cl, and Na indicate that sea salt particles over the entire particle size range sampled grow to cloud droplet size with high efficiencies. Approximately 80% or more of the masses of these elements on stages 3–6 are shifted toward stages 1 and 2. Approximately 50% of the masses of

I and Br concentrated in the afterfilter and stage 7 are shifted toward larger sizes.

The Al, Mn, and V size distributions indicated significant scavenging of only the largest particles collected by stages 2–4 of the Andersen impactor. Mass shifts from these stages toward cloud droplet sizes are generally <50% even for particles >1 μ in diameter. Small-particle masses are not significantly affected.

TABLE 3. In-Cloud Scavenging Efficiency Relative to Na

	% Efficiency for Element A*					
	$A = $ I	$A = $ Br	$A = $ Cl	$A = $ Al	$A = $ Mn	$A = $ V
CFR1/CFA1	160	90	140	5	20	10
CFR2/CFA2	170	130	160	5	20	20
CFSR2-1/CFA2	100	120	170	7	20	10
CFSR2-2/CFA2	120	140	160	6	10	20
CFSR2-3/CFA2	170	150	170	6	20	30
CFSR2-4/CFA2	150	150	170	7	20	30
CFSR2-5/CFA2	120	150	170	8	10	20
CFSR2-6/CFA2	130	140	160	7	20	40
Average %E	140	130	160	6	20	20

*Equation used to derive percent efficiency is $\%E = \dfrac{(A/\text{Na}) \text{ rain}}{(A/\text{Na}) \text{ total particulate}} \times 100$.

Comparisons of rain and total particulates. To get a general idea of the differences in behavior among the seven elements, we can compare the relative amount of each element found in the CF rain samples collected in the clouds with the relative amount of the element found there in aerosol and cloud droplet particles during the same time period. For this purpose it is necessary to compare the behavior of six of the elements to Na, since the total amounts of water condensing in the cloud or the degree of coalescence among cloud droplets is not known. Sodium is normally chosen as a standard of reference owing to its known association with sea salt and lack of gaseous phase. Therefore we will define a relative percentage rainout efficiency for element A as

$$\%E = \frac{(A/\text{Na}) \text{ rain}}{(A/\text{Na}) \text{ particulates}} \times 100$$

If $E = 100\%$, an element is rained out with the same efficiency as Na.

Table 3 summarizes the rainout efficiencies for I, Br, Cl, Al, Mn, and V relative to Na during the two particle- and rain-sampling periods. Samples labeled CFSR2-1–CFSR2-6 were collected sequentially during particle collection CFA2. It is interesting to note that I, Br, and Cl all have an apparent rainout efficiency greater than Na even though they are directly associated with the sea salt component of the aerosol. A possible explanation may be incorporation of gaseous phases of these elements during raindrop formation. Gaseous phases of I, Br, and Cl are all known to exist commonly in the marine atmosphere [*Duce,*

1969; *Moyers and Duce,* 1972a, b]. To see if any halogen gas adsorption takes place during cloud droplet formation, we can compare the halogen/Na ratios of the cloud droplets with these ratios for the entire particulate material present in clouds. Table 4 presents these comparisons for both in-cloud runs at station CF. The similarities in the halogen/Na ratios of the cloud droplets and total particulate matter suggest that no detectable incorporation of a gaseous phase has occurred during cloud droplet formation.

The rainout efficiencies of Al, Mn, and V shown in Table 3 seem very reproducible and are low in relation to Na. As we have seen, theory predicts this behavior for elements associated with silicate dusts such as Al and Mn. The similar behavior of V suggests that it may also be associated with the dust component of the aerosol.

SUMMARY AND CONCLUSIONS

Warm rain cloud physics theory suggests that hygroscopic sea salt particles should be more

TABLE 4. Cloud Droplet A/Na
Versus Total Particulate A/Na

Cloud Droplet and Aerosol Samples	Elemental Ratios A/Na		
	I/Na	Br/Na	Cl/Na
CFA1			
Total particles	0.0019	0.010	1.60
Cloud droplets	0.0017	0.0082	1.43
CFA2			
Total particles	0.0034	0.0081	1.33
Cloud droplets	0.0027	0.0069	1.38

easily nucleated than nonhygroscopic dust particles, this difference increasing with decreasing particle size.

The chemical composition of newly formed cloud droplets and rainfall should be equal to the total composition of aerosols acting as nuclei if there is no selective elemental absorption or volatilization.

In this work, differences in the behavior of elements associated with sea salt and soil dust aerosols during their transformation to cloud droplets and rain in orographic clouds were observed. These differences in scavenging efficiency may lead to different residence times in the atmosphere for the two groups of elements.

Comparisons of elemental particle size distributions in and out of clouds indicated that approximately 50–75% of the total masses of I, Br, Cl, and Na grew to become cloud droplets. Only a significant percentage of the largest particles or <25% of the total masses of Al, Mn, and V apparently associated with soil dust were observed to grow.

Comparisons of elemental ratios in simultaneously collected particles and rain at cloud base resulted in calculated scavenging efficiencies for six of the elements relative to Na. Owing to their association with hygroscopic sea salt particles and possible addition to rain via gas adsorption, scavenging efficiencies for I, Br, and Cl were 140, 130, and 160%, respectively, relative to Na.

Because of the lack of growth of smaller particles and lesser growth of larger soil dust particles to cloud droplet size relative to sea salt particles, Al, Mn, and V scavenging efficiencies relative to Na were 6, 20, and 20% respectively.

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