CHAPTER 3

Cloudwater and Aerosol Composition at Elevated Sites During the South Coast Air Quality Study

by

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Introduction

The Suggested Program Plan for SCAQS (Blumenthal et al., 1986) identifies the need to measure cloud and fog in the SoCAB in order to properly understand gas and aerosol processes. There is considerable evidence that the presence of fog or low cloud in the basin is associated with high SO² aerosol loadings and poor visibility in the SoCAB (Blumenthal et al., 1986, and references therein). Previous research sponsored by the CARB at Caltech has identified pre-existing aerosol as a major contributor to the acidity level in fog and cloud. Conversely, evaporated fog and cloud droplets are a major source of aerosol. The primary objectives of Caltech's participation in SCAQS are to assess the role that fog and cloud play in the formation of aerosol in the SoCAB (Objective 3, issue 1 of SCAQS) and to quantify the scavenging of precursor aerosol and gases during droplet formation. The extensive data available from SCAQS on meteorology, emissions, gas and aerosol concentrations will be used to evaluate cloud and fog droplet chemistry models. In this chapter we present the results of cloudwater and aerosol sampling at elevated sites in the SoCAB during SCAQS.

Methods

During the summer of 1987 cloudwater, aerosol and selected gases were sampled in conjunction with the South Coast Air Quality Study (SCAQS). During the period June 13 to July 17 sites were in operation at San Pedro Hill, Henninger Flats, and Kellogg Hill. A fourth site on Flint Peak, near Pasadena, was set up but could not be used because interference from FM transmitters located near the site disrupted the sampling equipment.

Site Descriptions

The San Pedro Hill site was located at a radar and communications facility operated by the U. S. Air Force and the Federal Aviation Administration. The elevation of the site is 450 m. The distance from the site to the ocean is 2.5 km. Los Angeles Harbor is 6 km east. The sampling equipment was placed at the edge of a flat grassy area. A steep hillside sloped away from the site, giving it unobstructed exposure from 70-270°. A SCAQS meteorological station was located a few hundred meters away from the sampling site.

Henninger Flats is at an elevation of 780 m, 7 km NE of Pasadena, and 45 km NE of the coastline. This site has been used in previous sampling programs (Waldman et al., 1985) and in a fog sampler intercomparison (Hering et al., 1987). A SCAQS meteorological station was also located at Henninger Flats.

Kellogg Hill is 38 km E of downtown Los Angeles at an elevation of 370 m. It lies 50 km NE of the coastline. The sampling equipment was located in a fenced enclosure adjacent to a small building that housed radio transmission equipment. The building partially obstructed the sampler when winds were from the south to west, which is the prevailing daytime wind. The site was unobstructed in the direction of the prevailing night-time winds. Construction activity near the site increased aerosol concentrations of soil dust during the daytime, and may have affected samples collected here.

Sampling Procedure

Each site was equipped with a Caltech Active Strand Collector with an automated fractionating sampler and a cloudwater sensor (see Chapter 1). The CASC collects droplets by inertial impaction on 510 μ m Teflon strands. The 50% collection efficiency

cutoff, predicted from impaction theory and based on droplet diameter, is 3.5 μ m. Also located at each site was an automated filter pack aerosol sampler. Teflon filters (Gelman Zefluor, 1 μ m pore size) were used to collect aerosol for inorganic analysis. HNO_{3(g)} was collected on a nylon filter (Gelman Nylasorb) placed behind one Teflon filter. NH_{3(g)} was collected on two oxalic acid impregnated glass fiber filters behind a second Teflon filter. The species that were measured in aerosol, gas and droplet phases are listed in Tables 1 and 2.

During the late afternoon or evening prior to expected cloud impaction, the samplers were cleaned by rinsing the collection strands, sample tubing, and reservoir with distilled, deionized water (DDH₂O). After rinsing, the strands were sprayed again with DDH₂O, which was collected in the fraction collector as a system blank. Rinsing and blank collection was repeated the following morning whether cloud was collected or not. Three sets of filters were loaded on the aerosol collector. A timer on the collector controlled the times that each filter set was run.

Some samples of cloudwater, fractionated by droplet size, were also collected and analyzed to determine whether any significant difference exists between the chemical composition of small vs. large cloudwater droplets. One of the CASC's was adapted for this purpose by adding an inlet containing four rows of eight 12.7 mm Teflon rods. These rods provide a 50% lower size cut of 16 μ m at the sampling velocity of 9 m/s. Each row covers 46% of the cross-sectional area. The four rows together sample 91.5% of the air passing through the collector. The sample collected by impaction on these rods is fed into a sample bottle. Droplets smaller than 15 μ m are collected inefficiently on the rods and pass through to be collected on the CASC strands in the main body of the collector.

Analytical Procedures

Samples were retrieved in the morning following a cloud event and transported to the lab at Caltech. The samples were weighed to determine their volume and the sample pH was measured with a Radiometer PHM82 pH meter using a combination electrode calibrated against pH 4 and 7 buffers. For selected samples small aliquots of the sample were removed and treated to stabilize reactive species. S(IV) was stabilized as the hydroxymethanesulfonate by adding buffered CH₂O (Dasgupta et al., 1981). CH₂O was stabilized with a solution of SO_3^{2-} (Dong and Dasgupta, 1987). A buffered solution of p—OH phenylacetic acid (POPA) and peroxidase was used to preserve peroxide (Lazrus et al., 1985). Carboxylic acids were preserved by addition of chloroform (Keene and Galloway, 1984). Carbonyls were derivatized with 2,4—dinitrophenylhydrazine in acidic solution (Grosjean and Wright, 1983).

The samples and preserved aliquots were stored in a refrigerator at 4°C until analysis. Major anions were determined by ion chromatography with a Dionex AS4 or AS4A separator column and a micromembrane suppressor. The eluent was 2.8 mM $HCO_3^{-}/2.2 \text{ mM } CO_3^{2^-}$. The metallic cations were determined by atomic absorption spectrophotometry. An air/acetylene flame was used for Na⁺ and K⁺; N₂O/acetylene was used for Ca²⁺ and Mg²⁺ to minimize interferences. NH₄⁺ was determined by flow injection analysis employing the indophenol blue method.

The stabilized CH_2O was determined by a modification of the Nash method for use with an autoanalyzer (Dong and Dasgupta, 1987) Hydrogen peroxide was added to eliminate S(IV), which interferes by forming an adduct with CH_2O . The absorbance of the colored product was measured at 412 nm. S(IV) was analyzed by the pararosaniline method (Dasgupta et al., 1980) adapted for flow injection analysis. Peroxide was determined from the flourescence of the POPA enzyme solution (Lazrus et al., 1985). The method is sensitive to H_2O_2 and some organic peroxides, however, the significantly lower

Henry's law coefficients of CH_3OOH and peroxoacetic acid suggest that they will not be important in fog and cloud water (Lazrus et al.,1985). Carboxylic acids were determined by ion exclusion chromatography (Dionex ICE-AS1) with dilute HCl as eluent.

The Teflon and oxalic acid impregnated glass fiber filters were extracted in distilled deionized water (DDH₂O) on a shaker table. A small volume of ethanol was added to the filter prior to extraction to more effectively wet the filter surface. The nylon filters were extracted in $HCO_3^{-}/CO_3^{2^{-}}$ IC eluent. Composition of the extracts was determined by the same procedures used for the fogwater samples, with the exception of additional buffer in the complexing reagent and oxalic acid in the rinse solution of the ammonia analysis to account for the effect of the oxalic acid.

The precision and accuracy of the measurements was determined from the mean and standard deviation of repeated analyses of the same standard (Standard Methods). Detection limits were estimated from analyses of blanks (or the lowest standard). The stated detection limit is defined as the concentration giving a signal 3 times the blank (or low standard) standard deviation. The expected precision, accuracy and detection limits for the major species measured are listed in Table 3. For gas—phase species the variance of the flow measurements is included in the estimates of precision and detection limit.

Results and Discussion

Cloudwater and Aerosol Composition

During the summer portion of SCAQS, stratus clouds frequently impacted the coastal slopes near San Pedro. This is reflected in the fact that over 240 samples were collected at San Pedro Hill during the study. Clouds were present at this site primarily during the night, although some events lasted until mid-day. On several occasions, the stratus extended as far inland as the slopes of the San Gabriel Mountains and was at the right elevation to be collected at Henninger Flats, where 76 samples were obtained. Stratus were rarely observed at the Kellogg Hill site during this study, where only a couple

of samples were obtained. The compositional data for the San Pedro Hill and Henninger Flats samples are presented in Tables 4 and 5.

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Samples at both San Pedro Hill and Henninger Flats were consistently acidic. Sample pH values at San Pedro Hill varied from 2.4 to 5.0, while those at Henninger Flats ranged from 2.6 to 4.8. The arithmetic average of the pH at each site was 3.25. Frequency distributions of the sample pH for each site are presented in Figures 2 and 3.

The composition of the cloudwater sampled at both sites was usually dominated by NO_3^- , SO_4^{2-} , NH_4^+ , and H^+ . In some samples Na⁺ and Cl⁻ were also found to be important contributors. Concentrations of Na⁺ and Cl⁻ were generally both observed at higher levels in the San Pedro Hill cloudwater, consistent with its proximity to the ocean. NH_4^+ concentrations averaged higher at Henninger Flats, while NO_3^- concentrations averaged approximately 1200 μ eq/l at both sites. SO_4^{2-} concentrations averaged one—third higher at San Pedro Hill (917 μ eq/l) than at Henninger Flats (689 μ eq/l).

Most of the samples obtained at the two sites were collected during non-overlapping time periods. Often the stratus clouds would impact one site on a given day and not the other. At other times, the clouds were observed to impact the hillside at San Pedro during the period shortly after midnight, while impaction at Henninger Flats did not begin until a few hours later. By this time, clouds were no longer intercepting the slopes of San Pedro Hill. The only event with simultaneous collection at both sites was on July 16. This event, which was associated with drizzle and rainfall in the L.A. Basin, was one of the most extended observed during the study. The sampling capacity of the autosampler carousel was exceeded at both sites at approximately 2300 on the 16th. The event was also one of the least acidic observed at San Pedro Hill. The cloudwater pH there climbed as high as 4.98 during the event, while all of the samples collected at Henninger Flats had a pH less than 4. Like H⁺ concentrations, concentrations of NO_3^- , $SO_4^{2^-}$, and NH_4^+ were also much higher at Henninger Flats during this event than they were at San Pedro Hill.

The combination of the high sampling rate of the CASC (the CASC has sampled at rates of up to 8.5 ml min⁻¹ in past field studies) and the use of the autosampler enabled us to collect samples with a very fine time resolution. During some events, 60 ml samples were collected at San Pedro Hill as quickly as one every 10 minutes. This time resolution enables an examination of rapid fluctuations in the cloudwater composition as a function of changes in wind direction or liquid water content. An example of this resolution is depicted in Figure 4. The diagrams in this figure illustrate the changes in the sample pH, the concentrations of the measured ions, and the estimated liquid water content (based on the collection rate and the theoretical sampling efficiency of the CASC), as a function of time, observed at San Pedro Hill for the period from 0100 to 0315 on June 25, 1987. In just over two hours, the observed levels of all these parameters were seen to change dramatically. Concentrations of all measured species were observed to drop by at least a factor of two between 0130 and 0230. These drops coincided with a doubling of the liquid water content, suggesting that the concentration changes were largely due to dilution. When the liquid water content began to fall after 0230, however, not all of the species' concentrations increased. Levels of SO_4^{2-} , NO_3^{-} , and H^+ did rise, but Na^+ , Cl^- , Ca^{2+} , and Mg²⁺ concentrations remained steady or declined. Analysis of meteorological data from San Pedro Hill, which is part of the SCAQS data set, will enable us to correlate changes in chemical composition with meteorological variables.

The aerosol and gas—phase data for San Pedro Hill, Henninger Flats, and Kellogg Hill are presented in Tables 6 through 8. The major species present are NH_4^+ , NO_3^- , and SO_4^{2-} . Na⁺ and Cl⁻ are also major components in some samples, particularly at the San Pedro Hill site in the daytime samples, which were collected during onshore flow conditions. The concentration of NH_4^+ is greater at the inland sites, Henninger Flats and Kellogg Hill, than at San Pedro Hill. During the daytime concentrations of $HNO_{3(g)}$ are quite high; they often equal or exceed the aerosol NO_3^- concentration. The gas—phase NH_3

concentrations are fairly low at Hennigner Flats and San Pedro Hill. Kellogg Hill, which is the closest site to the dairy feedlots in Chino, has the highest $NH_{3(g)}$ concentrations.

Blank Concentrations

In order to determine the efficacy of our collector cleaning procedure the collection strands in the CASC were sprayed with distilled water to generate blanks. The concentrations of major ions in the collector blanks are presented in Table 9. In general the blank concentrations are an order of magnitude less than the measured concentrations of the samples, although there were a few cases of fairly high concentrations in the blanks. Rinsing the strands effectively removed material from the collector; analysis of successive blanks indicates decreased concentrations in the second rinse compared to the first.

Size-fractionated Samples

Clouds began impacting the San Pedro Hill site at approximately 1900 on July 14 and remained there until approximately 1300 on July 15. The event was sampled by the CASC and the autosampler from 1900 until approximately midnight, when the last bottle in the carousel was used. Manual operation of the CASC, with the size-fractionating inlet installed, began at 0400. Samples were collected at half-hour intervals until noon on the 15th. A final one-hour unfractionated sample was collected from 1200 to 1300. The size-fractionating inlet, as discussed earlier, is used to collect the larger droplets in the cloud (those with diameters greater than about 16 μ m), leaving the smaller droplets to be collected on the CASC strands. Figure 5 depicts the fraction of the droplet-size spectrum collected by each portion of the sampler for three different liquid water contents. The droplet-size spectra are typical representations based on the work of Best (1951a). The fraction of the liquid water sampled by each part of the collector is calculated based on

impaction theory for droplets on a cylinder. At low liquid water contents, where a higher proportion of the water is typically contained in smaller droplets, most of the droplets will pass through the fractionating inlet and collect on the CASC strands. At higher liquid water contents, however, the preponderance of large droplets will shift the bulk of the collected sample volume to the fractionating inlet. As shown in the figure, considerable overlap in the portions of the droplet spectrum sampled by each part of the collector are expected. Any differences seen in the composition of the two sample fractions, therefore, should be considered as a lower bound on the differences present between more sharply divided portions of the spectrum.

The chemical compositions of the samples collected using the size-fractionating inlet are displayed in Table 10. The samples have been labeled "f" for those fractions collected on the large rods in the front of the collector, and "b" for those collected on the CASC strands in the back. The volume weighted average concentration of each species, obtained by mathematically combining the two fractions for each sampling period, is also presented. The two fraction volumes collected during each sampling interval are compared in Figure 6. The ratio of volumes collected in each fraction was on the order of two or three to one, in favor of the fraction collected on the large rods, for all sampling intervals. During several periods the cloud grew dense enough to produce a light drizzle. Consistent with the predictions in Figure 5, the ratio of volumes collected by each fraction was higher during the periods when the liquid water content was higher.

Comparisons of the ion concentrations in the two fractions collected during each interval suggest that there is a large difference between the average composition of the smaller droplets and that of the larger droplets. Figures 6 through 8 illustrate the case for each of the species measured. For every interval sampled, the concentration of Na⁺ and Mg²⁺ in the small droplet fraction was observed to be higher than in the large droplet fraction. With the exception of one very low concentration sample, the same was true for Ca²⁺. Concentrations of SO₄²⁻, NO₃⁻, NH₄⁺, and H⁺ were almost always higher in the

small droplet fraction (the fact that they were lower in this fraction for the first couple of samples may be due to dilution by residual rinsewater on the CASC strands). Concentrations of Cl⁻ showed no consistent preference for one fraction over the other, and were generally at similar levels in both fractions. The composition of the fractionated sample collected between 0600 and 0630 is depicted in a pie diagram (Figure 9). In addition to the differences discussed above, this figure illustrates that the total of the ionic species concentrations is much higher in the smaller droplets, on average, than in the larger ones.

Similar results were observed when comparing samples collected by the Caltech Rotating Arm Collector and the CASC at a coastal site in the Santa Barbara Channel. The RAC (Jacob et al., 1984) which has a 50% lower size cut, based on droplet diameter, of about 20 μ m, collected samples with higher concentrations of Na⁺, Ca²⁺, and Mg²⁺ than were observed in samples collected by the CASC (Collett et al., 1988). The difference in the concentrations was attributed to differences in the portions of the droplet-size spectrum sampled by the two collectors. The CASC, with its theoretical lower size cut of 3.5 μ m, collects almost all of the liquid water contained in cloud droplets, while the higher size cut of the RAC allows it to only collect the larger droplets efficiently. Typically, larger condensation nuclei lead to the formation of larger cloud droplets in the lower portions of a cloud not subjected to significant horizontal entrainment of dry air (Best, 1951b; Mason and Chien, 1962; Hudson, 1984). Since sea salt and soil dust are found to reside in the larger end of the aerosol size spectrum (Seinfeld, 1986), the elements found predominantly in these types of particles (e.g. Cl, Na, Mg, and Ca) will reside there as well. Therefore, as cloud droplets form on the available aerosol nuclei and grow by condensation, it is expected that these elements will be found predominantly in the upper end of the droplet size spectrum.

During the same study, no preference was shown by either collector for higher concentrations of NO_3^- , SO_4^{2-} , NH_4^+ , or Cl⁻. NO_3^- , SO_4^{2-} , and NH_4^+ , which make their

way into the aerosol phase largely by gas-to-particle conversion processes, are found to reside largely in smaller aerosol particles (Seinfeld, 1986). One might expect then that smaller cloud droplets would be enriched in these ions relative to their larger counterparts. The differences might not be so readily apparent in the droplet phase, however, since all three species may also be introduced by absorption of gas phase precursors, coupled with oxidation of S(IV) to S(VI) in the case of SO_4^{2-} . Absorption of $NH_{3(g)}$, and particularly of $HNO_{3(g)}$, was seen to play an important role in determining the composition of cloudwater at the study site (Collett et al., 1988). Since gas phase absorption can make important contributions to the composition of all droplet sizes, it may tend to mask the initial chemical signature given to the droplets by their condensation nuclei.

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Observing differences in the chemical composition of small vs. large droplets in the present study is facilitated by the way the droplet size spectrum has been split. While there is still considerable overlap in the portions of the size spectrum collected by the large rods and the small CASC strands, there is more difference than in the Santa Barbara Channel comparison, where the average composition of the large droplets was being compared to the average composition of the whole spectrum. This may at least partially explain why differences in the concentrations of NO_3^- , SO_4^{2-} , H⁺, and NH_4^+ are evident this time.

The distribution of Cl⁻ between large and small droplets is somewhat different from the other ions discussed, being found at roughly equivalent concentrations in each fraction. Cl⁻ is initially associated with Na⁺ in sea-salt aerosol. If it remained there until nucleation of the aerosol within the cloud, we would expect to see Cl⁻ concentrations higher in the large droplets, as was observed for Na⁺. This clearly is not the case, suggesting the possibility that Cl⁻ was introduced into the droplets in another form. Aerosol samples collected on the afternoon of July 14 at San Pedro Hill show substantial concentrations of Na⁺, but essentially no Cl⁻ (see Table 6). High HNO_{3(g)} concentrations also observed during this period could have resulted in acid exchange of HCl_(g) for HNO_{3(g)} in the sea

salt aerosol. Introduction of Cl⁻ to the cloudwater droplets that evening as HCl(g) would have produced the type of uniform distribution among all droplet sizes that was observed.

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Table 1. Analysis of liquid samples.

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Method

Combination Electrode pН Ion chromatography - HCO_3^{-}/CO_3^{2-} eluent C1-NO₃⁻ S042-NH4⁺ Indophenol blue - Flow Injection Analysis Na* Flame Atomic Absorption Spectrometry K* Ca²⁺ Mg²⁺ S(IV) Pararosaniline - FIA CH_2O Nash Method C1 - C6 Carbonyls DNPH Derivative - HPLC Ion Chromatography - B₄07²⁻ eluent Carboxylic Acids Ion Exclusion Chromatography - HCl eluent H_2O_2 POPA - flourescent dimer

Table 2. Analysis of gas and aerosol samples.



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Table 3.

PRECISION, ACCURACY, AND MINIMUM DETECTION LIMIT

Aqueous Phase

Species	nominal conc.	Acc.	Prec.	RSD	MDL
	μ N	μN	μN	*	μ N
NH 4 *	20	0.28	0.58	2.92	1.75
Na ⁺	10.9	0.13	0.45	4.15	1.36
Ca ²⁺	12.5	-0.42	0.87	6.97	2.61
Mg ²⁺	10.25	0.30	0.48	4.66	1.43
C1-	20	0.26	2.04	10.20	6.12
NO ₃ ⁻	20	0.11	0.92	4.60	2.76
S042-	20	0.20	0.86	4.29	2.57
C1-	2	0.87	2.22	111.07	6.66
NO3 -	2	0.31	0.35	17.71	1.06
S 042	2	0.26	0.38	19.13	1.15

Gas Phase (4 hour run)

Species	nominal conc.	Acc.	Prec.	RSD	· MDL
·	neq m ⁻³	neq m ⁻³	neg m ⁻³	*	neq/m ⁻³
NH 4 +	69.42	0.96	2.90	4.18	8.70
Na ⁺	37.84	0.44	1.95	5.16	5.86
Ca ²⁺	43.39	-1.46	3.28	7.56	9.84
Mg ²⁺	35.58	1.04	1.99	5.61	5.98
C1-	69.42	0.90	7.40	10.65	22.19
NO ₃ -	69.42	0.38	3.83	5.52	11.49
S042-	69.42	0.69	3.66	5.27	10.97
NH ₃	69.42	0.96	2.90	4.18	8.70
HNO ₃	69.42	0.38	3.83	5.52	3.69
C1-	6.94	3.02	7.71	111.07	23.13
NO ₃ -	6.94	1.09	1.23	17.71	3.69
S042-	6.94	0.91	1.33	19.13	3.98

Table 4 San Pedro Hill Cloudwater Data

San Pedro Hill Cloudwater Data

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Date	Sec	Date	Seq	Start	Stop	Vo1	рH	Ha+	K+	MH4+	Ca2+	Hg2+	"Fo"	C1-	NO3-	S042-	CH20	H202	NFo	HAC	LHC
						(11)		(uN)	(uN)	(UN)	(uN)	(uli)	(uli)	(uN)	(UN)	(uli)	(ull)	(UII)	(ull)	(ull)	al/s3
05/13	1	05/13	1	20:43	21:22	49	3.28	516	21	471	88	119	161	474	\$55	\$78					0.070
06/13	2	05/13	2	21:22	21:52	53	3.19	369	14	377	52	166	144	407	743	661					0.099
06/13	3	05/13	3	21:52	23:19	68	3.18	592	21	496	74	178	175	558	904	781					0.044
06/15	4	05/:3	- 4	23:19	QC:20	62	3.08	587	22	588	70	184	171	605	\$57	\$24					C.057
06/14	5	05/14	5	00:20	00:56	48	2.89	688	24	562	80	214	147	681	1321	1215					0.075
06/14	6	06/14	6	00:56	01:41	56	2.80	647	22	555	71	190	139	587	1608	1329		,			0.070
06/14	7	05/14	1	01:41	02:45	57	2.85	705	24	\$25	78	202	172	762	1554	1296					9.050
05/14	8	06/14	8	02:45	03:13	47	3.07	731	23	495	75	214	133	771	962	895					0.094
06/14	9	05/14	9	03:13	03:46	51	3.09	440	15	376	43	119	118	465	796	722					8.087
06/14	10	06/14	10	03:46	04:09	42	3.00	361	13	402	38	\$5	116	400	893	\$72					8.102
06/14	11	05/14	11	04:09	04:31	44	3.06	265	10	375	27	64	113	327	819	736					9.111
05/14	12	06/14	12	04:31	04:59	53	3.12	149	5	323	15	47	119	247	702	574					0.105
35/14	13	06/14	13	04:59	05:40	34	2.98	263	11	477	27	82	141	400	993	870					0.047
06/14	14	05/14	14	05:40	06:16	34	2.91	538	20	64 8	55	155	169	599	1249	1125					8.052
06/14	15	05/14	15	05:16	07:13	18	2.82	734	28	744	77	208	181	787	1569	1407					9.017
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06/15	4	06/15	4	04:48	05:22	36	3	3796		1237	450	731	163	1701	2137	1125	25				0.055
06/19	5	86/19	5	05:22	06:02	36	3	3405		\$27	318	634	158	1672	1503	1131	31				8 051
06/19	6	06/19	ŝ	06-02	C7 - DA	26	3	3258		665	281	593	130	1654	1281	978	27				0 023
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06/20	1	06/20	۱	00-29	01-44	50	3 11	2574		555	768	488	150	1430	1402	1044	31				0 044
06/20	;	66/20	,	01-44	02.28	62	1 14	1768		- 605	187	410	126	1784	1025	1046	24				0 070
06/20	÷	06/20		02.28	02.25	22	3 26	2318		485	173		119	1370	916	783					0.053
85/20	1	06/20		02.20	04.02	65	3 35	1977		125		100	28	14.71	E18	740					8 004
06/20	Ē	06/20	ī	D4 02	04.02	90 90	2.35	1740		141		411	14.0	1476	248	1929					0.030
86/20	ŝ	00/20	-	54.46	05.05		3.33			617	777	716	176	1966	477	1292					0.000
	•		•		03.03	•	3.20			917	414	113	114	1190	• * *	1230					9.920
85 / 21		06/21		00.14	AD - 55	22	1.48	805		785	147	963		775	815	447	54				
86/21		06/01		00.14	01.37	22	3 6 3	\$33		910	102	100	97	195 801	477	950	19				0.003 A 167
06/21	10	06/21	10	ñ1.17	01.42	19	9 85	706		160	78	182	81	821	997	313	16				0.000
99/2	10	00/21	16	V 1:17	91:44	14	ə. əə	100		103	(3)	184		8 97	311	312					8.849
86 /22	,	06 / 23	•	60.94	A1.12		3 43	1967		953	-		186		1814	205					
96/22	, ,	ND/22		01.13	01213	60	3.83	1231		911	243	311	100		819	30/	11				0.970
00/22	4	00/22		01:13	01:01		3.37	9 4		311		110	193	440	341	448	12				0.124
00/22	3	00/22	5	01:41	02:17		3.31	231		321	41	75	183	333	479	341	15				Q. US:
¥0/22	- 4 - E	90/22	4	02:17	02:50	• • • •	3.54	24.5		333	31	• • • •		289	436	354	14				9.103
06/22		06/22	,	92:50	03:27	\$2	3.36	206		376	27	53	100	257	458	391	15				8.894
00/22	•	U0/22	0	03:27	U4:15	02	3.20	214		60) 	29	×	121	303	371	405					U.075
06/22		06/22	1	04:13	05:17	50	3.09	342		640	51		145	436	852	865	20				0.052
UD/22		06/22		05:1/	05:34	13	2.89	002		1025	109	108	510	989	1447	1121					0.042
06/22	· 9	06/22	3	06:00	07:16	50	2.44	\$78		1126	113	151	202	611	1632	1478	27				0.944
06/22	1	06/22	1	23:18	23:58	66	3.32	285		371	15	12	17	334	- 544	450					0.09
06/22	2	00/22	2	23:58	00:20	62	3.44	242		349		70	102	200	479	370					0.12
00/23	د •	00/23	3	00:25	01:05	12	3.31	212		354	57	60	97	263	537	455					0.09
06/23		06/23	4	01:05	C1:39	52	3.36	250		348	53		76	307	473	391					0.10
06/23	5	06/23	5	01:39	02:15	52	3.49	747		309		66	76	322	334	284					0.10
06/23	6	08/23	6	02:15	02:44	51	3.51	255		270	39	57	57	317	362	333					0.1Z
36/23	1	05/23	1	02:44	03:07	52	3.54	200		231	30	54	55	262	238	285					0.15
05/23	t	05/23	1	03:07	03:31	52	3.48	196		229	30	54	56	259	322	318					0.15
06/23	9	06/23	9	03:31	03:55	62	3.54	174		234	24	47	58	231	295	269					0.15
06/23	10	06/23	10	03:55	04:18	62	3.50	181		264	27	41	50	226	349	320					0.15
06/23	11	06/23	11	04 : 18	04:49	62	3.50	162		295	24	42	71	290	424	352					0.11
06/23	12	06/23	12	04:49	05:16	62	3.62	141		280	18	38	25	196	271	224					0.13

San Pedro Hill Cloudwater Data

Date	Sec	- Hate	266	Start	Stop	¥0)	pH.	Ha+	X +	MK4 +	Ca2+	R02+	"Fo"	C1-	103-	5042-	CH20	NZ02	MFo	HÁC LHC	
						(1)		(ull)	(uli)	(ull)	(ull)	(ull)	(ull)	(uli)	(ull)	(ull)	(uill)	(uii t)	(ull)	(uff) #1/#3)
06/77	17	n s /33	12	55.16	65.41	61	3 55	126			10	27		180	320	983				* ••	
06/23	14	06/23	14	05.41	06-07	62	3 55	193		202	17	31	24	192	320	141					
06/23	15	06/23	15	06-07	05-29	\$2	3 58	131		733	20	35		177	107	277				0.13)
06/23	16	06/23	16	06:29	06-54	62	3 58	130		226	21	36	27	184	297	269				6 16	
06/23	17	06/23	17	06:54	07:20	62	3.52	148		272	23	40	36	203	323	213				0.13	i
06/23	18	05/25	18	07:20	07:43	52	3.47	169		308	28	45	43	218	388	371				0,15	ŝ
05/23	19	06/23	19	07:43	08:05	62	3.43	171		293	30	47	44	225	392	358				0.15	
06/23	20	06/23	20	05:05	10:27	62	3.15	452		528	95	131	104	505	1024	506				0.02	2
05/23	1	06/23	1	19:55	20:47	30	2.53	2941		1659	527	561	586	1574	5623	3246	35			0.03	
06/23	1	06/23	- 1	22:30	23:18	61	2.64	3148		1510	56C	601	497	1548	5240	284.5	26			0.07	
96/23	9	06/23	9	23:18	00:21	53	2.57	29 53		1725	505	577	470	1613	5287	2934	22			0.05	
06/24	10	06/24	10	00:36	01:09	\$ 1	2.52	1289		1520	240	313	259	824	3617	2291	25			0.10)
96/24	11	06/24	11	01:09	01:33	6 1	2.82	44.5		789	78	134	147	492	1770	1341	18			0,14	
06/24	12	05/24	12	01:33	02:04	61	2.79	276		726	45	\$2	116	393	1586	1329	19	-		0.11	I
06/24	13	06/24	13	02:04	02:31	61	2.81	236		143	34	- 64	133	339	1667	1254	23			8.13	I
06/24	- 14	06/24	14	02:31	03:03	§ 1	2.71	225		1443	- 41	- 58	133	346	2171	1433	30			\$.11	1
05/24	15	05/24	15	03:03	03:38	61	2.71	295		2147	55	- #	152	456	2715	2053	31			0.10	
06/24	16	05/24	16	03:38	04:12	61	2.75	238		2859	60		160	505	2757	2501	49			0.10	1
06/24	17	06/24	17	04:12	04:42	61	2.75	284		2441	41	79	157	475	2518	2205	46			Q.11	
05/24	18	05/24	10	04:42	05:03	51	2.85	35C		1465	50		115	441	1849	1561	31			0.16	į
00/24	19	08/24	19	05:03	85:17	62	7.54	248		399	30	65	4	360	1291	1115	29			0.25	
00/24	20	V0/24	100	reser	7015	90	2.84	185		1748	33	57	113	203	2073	1538					
80/24	490	08/24	41	- TESET"	01.04	84	2.83	134		1910		24	118	318	2108	138/	41				
46/24 66/94	- 21	06/24	- 41	00:41	07.66	90 60	4.81	185		2980	30 45	20	130	490	5833	1853	45			0,12	!
NO/24 86/24	22	06/24	- 44 - 92	07.20	01:30	80 60	4.18	230		2130	43		184	337	2340	1931	43			0.07	,
00/24	24	90/24 RE/94	23	NI130	08.33	90 90	4.19	404		1840	*0	10	131	330	2000	1240	80 £1			9,13	j I
00/24	24	40/24	- 24	00:21	09:32 Ng.26	2V 81	5.10	453		1310	88 102	194	149	603	2100	1321	31			Ų. IU A. 14	i L
06/24	26	86/24	25	00.02	00.30	61 61	5.11	973	16	467	107	139		474	1676	1390				0.14	,
06/74	27	06/24	27	00.14	09.33	61	2 42	185	16	505			80	477	1434	1196	20			· D 16	
05/24	28	05/24	28	09.33	10-42	60	2 65	1790	- 55	1457		377	979	1365	1621	2358	ีน			8.65	
05/24	29	05/74	29	10:42	11-19	17	2 60	1803	59	1341		406	240	1395	3602	2452	-			8.63	,
•-, ••	•••	•••	•••			•••			••				***							•.••	•
06/24	1	05/24	1	19:39	23:02	11	2.66	1518	307	9383	1143	1151	393	2096	8131	5796				8.00	1
06/25	3	06/25	3	80:37	01:04	\$6	2.63	594	27	1855	200	135	211	400	3658	2438	35			0.14	l
96/25	4	06/25	4	01:04	01:33	65	2.57	\$10	30	1915	239	173	210	488	3481	2422	37			0.13	J
06/25	5	06/25	5	01:33	01:53	67	2.73	708	25	1242	193	159	151	440	2646	1815	31			8.19	}
06/25	5	06/25	- 6	01:53	02:07	67	2.72	395	- 14	724	77	\$1	\$2	270	1471	1105	24			9.27	
08/25	1	06/25	1	02:07	02:20	67	3.09	244	11	470	43	53	46	176	832	662	15			0.29	l
06/25	1	06/25	1	02:20	02:31	57	3.11	205		410	38	50	33	157	145	711	- 16			0.34	
06/25	9	06/25	9	02:31	92:42	67	3.11	270	10	429	55	6 1	- 44	184	\$70	741	17			0.34	1
06/25	10	06/25	10	02:42	02:56	67	3.94	177	1	509	30	40	34	132	\$9 1	660	30			0.27	
06/25	11	06/25	11	02:56	03:12	67	2.87	158	1	1078	32	37	101	174	1805	1120	30			0.23	\$
06/25	12	06/25	12	03:12	03:14	10	2.88	142	7	1031	33	37	\$2	162	1837	1995				0.29	I.
06/25	13	06/25	13	03:16	03:41	- 64	2.74	171	10	1270	36	44	132	230	2583	1528	14			0.15	i
06/25	14	06/25	14	03:45	03:53	18	2.66	273	15	1753	57	66	146	276	3270	1895				0,13	1
06/25	15	06/25	15	03:57	04 : 06	26	2.67	289	13	1708	52	66	132	265	3020	1828	28			0.16	1
06/25	16	06/25	- 16	04:09	04:17	20	2.65	315	14	1631	50	72	152	281	4000	1854	.			0,14	
06/25	11	00/25	17	04 : ZQ	U4 : 27	Z3	2.70	224	13	1064	55	65	140	272	Z823	1588	72			0,18	ļ
V0/25	18	06/25	18	94:30	04:37	21	Z.73	285	13	1423	54	59	135	295	7490	1702	Z3			0.17	
00/25	19	00/25	19	04:39	94:45	13	2.71	434	16	1735		#6	148	364	2888	Z106				0.12	:
	n	- UN 7 2 4	20	114 • # #	ua - 57	10	2 87	- NA 1	76	1714	117	104	147	386	2020	7186				1 14	

San Pedro Hill Cloudwater Data

Date	Sec	Date	Seq	Start	Stop	Vol	pH	Na+	K+	9814+	Ca2+	Mg2+	"Fo"	¢1-	W03-	S042-	CH20	H202	NFo	HAC LHC
						(#1)		(uli)	(uN)	(uN)	(uli)	(uN)	(ull)	(uli)	(uN)	(uli)	(uli)	(ull)	(ulli)	(ull) m1/m3
06/25	21	06/25	21	D4 : 55	05:04	19	2.65	511	22	1684	115	110	152	407	3233	2222				0.12
06/25	22A	06/25	22A	reser	voir	\$7	2.74	512	18	1\$55	90	91	135	476	2575	2234	25			
06/25	22B	35/25	228	reser	voir	44	2.75	520	18	1736	88	96	127	468	2362	2958	24			
06/25	23	06/25	23	05:17	07:33	17	2.54	1266	44	2373	270	256	215	\$38	3435	3229				0.01
06/25	1	06/26	1	01:48	02:08	33	2.99	315	19	1979	154	86	129	213	2462	1528				0.09
06/26	2	06/25	2	02:08	02:19	39	3.26	113	1	1704	41	30	\$8	89	1445	1045				0.20
06/26	3	06/26	3	92:19	02:30	40	3.33	\$7	- 4	906	19	18	26	77	\$22	713				0.20
06/25	4	06/26	4	02:30	02:57	37	3.30	158	1	1035	43	39	33	- 14	1054	762				0.08
06/25	5	06/26	- 5	02:57	83:06	33	3.27	83	5	1363	22	24	23	43	\$12	645				0.20
06/26	6	06/26	6	C3:06	03:15	35	3.36	49	4	1124	13	16	26	51	1015	747				0.23
06/25	7	06/26	7	03:15	03:24	32	3.39	67	5	1119	23	23	49	88	1095	\$10				9.20
06/26	1	06/26	1	03:24	03:39	\$4	3.42	103	1	1415	22	26	58	\$7	1113	\$23				0.24
06/25	•	06/26	9	03:39	03:53	68	3.48	41	4	879	17	16	39	55	\$14	590				0.27
96/26	10	06/26	10	03:53	04:04	39	3.48	31	5	593	19	24	25	106	572	534				0.20
06/26	11	06/25	11	04:04	94:24	\$6	3.51	114	1	479	27	30	22	- 54	489	461				8.19
96/26	12	06/25	12	04:24	04:43	- 65	3.47	59	4	423	12	17	23	58	454	387				8.19
06/25	13	06/25	13	04:43	05:00	69	3.43	41	3	478	11	13	29	51	560	393				0.23
06/26	14	05/25	- 14	05:00	05:14	64	3.40	28	3	585		11	35	45	718	482			•	0.26
06/26	15	06/25	15	05:14	05:27	63	3.38	33	3	709		10	46	47	750	496				9.27
95/25	16	06/25	15	05:27	05:45	69	3.24	45	3	\$43	10	13		- 64	1951	680				8.21
06/25	17	06/26	- 17	05:45	05:58	68	3.28	36	3	886		10	58	78	· \$55	643				0.29
06/26	18	06/26	18	05:58	06:12	69	3.28	31	3	787		11	\$5	\$5	965	663				0.27
05/25	19	06/25	19	06:12	06:25	63	3.26	35	3	1025	13	12	70	56	1055	748				8.25
06/28	ZUA	06/26	ZUA	reser	VOIR	65	3.06	165		1177	38	43	123	195	1625	1553				
06/26	208	06/26	208	reser	voir	65	3.90	160	10	1818	45	43	146	223	1998	1971				
07/01	1	07/01	1	20:34	21:57	29	3.18	\$12		638	215	24 \$	88	745	1138	836				. 02
07/01	4	07/01	4	22:07	22:10	1	3.15	\$18		613	159	175	79	544	936	775				0.15
07/01		07/01	. 1	22:33	22:39	13	3.15	386		522	97	108	67	366	722	\$58				8.12
07/01	10	07/01	10	22:51	22:56	10	3.18	335		496	79	90	52	339	682	585				1.11
07/01	11	07/01	11	22:59	23:08	17	3.19	372		565	88	19	66	378	704	666				. 8.11
07/01	13	07/01	13	23:19	23:24	11	3.23	348		545	80	20	54	351	605	609				Q. 12
07/01	20	07/01	20	reser	voir			38 5		242	57	50	46	453	401	435.				
07/02	1	07/02	1	20:00	21:10	60	3.19	528		494	\$5	133	109	510	713	836				8.05
07/02	2	07/02	2	21:10	22:46	34	3.30	379		394	75	93	- 11	400	535	492				8.02
07/03	3	07/03	3	00:05	00:55	62	2.91	683		2132	286	202	221	438	3206	1546				8.07
07/03	4	07/03	- 4	OC:55	01:32	\$1	2.99	345		2309	125	96	210	327	2325	1464				0.09
07/03	5	07/03	5	01:32	02:48	52	2.60	515		2770	209	158	236	532	4131	2562				0.94
07/03	6	07/03	6	02:51	03:50	61	2.47	487		2568	167	149	271	562	4656	3028				8.06
07/03	7	07/03	7	03:50	04 : 35	62	2.51	392		2156	118	110	219	488	3278	2305				0.08
07/03	8	07/03		04:35	05:12	60	2.67	377		1725	\$8	105	179	454	2748	1903				0.09
07/03	9	07/03	9	05:12	05:46	61	2.54	405		1557	100	115	176	479	2994	1959				0.10
07/03	10	07/03	10	05:46	06:22	60	2.63	308		1682	72	83	180	440	2924	2030				0.09
07/03	11	07/03	11	06:22	06:53	59	2.61	315		1195	71	83	159	365	2874	1691				0.11
07/03	12	07/03	12	06:53	07:25	58	2.58	355		1037	82	15	180	381	2973	1687				0.10
07/03	13	07/03	13	07:25	08:15	58	2.48	446		1425	110	124	256	491	3823	2340				0.06
07/03	14	07/03	14	08:15	05:08	20	2.42	672		1693	186	192	298	\$56	4631	2957				0.02
07/07	1	07/07	1	00:54	01:29	\$6	3.33	647		1108	127	145	135	582	1301	810	20	4		0.11
07/07	2	07/07	2	01:29	01:47	57	3.31	388		544	61	88	84	405	\$12	588	15	1		0.18
07/07	3	07/07	3	01:47	02:02	58	3.34	235		320	28	53	72	284	554	455	14	18		0.21

Table 4 San Pedro Hill Cloudwater Data (continued)

San Pedro Hill Cloudwater Data

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Date	Sec	Dece	Sec	Start	Stop	Vol	pH	Na+	K+	M14+	Ca2+	H 52+	"Fo"	¢1-	1103-	\$042-	CH20	W202	HFo	HAC	LHC
						(1)		(uN)	(ull)	(uli)	(uli)	(illi)	(uli)	(ull)	(uN)	(ull)	(ull)	(ull)	(JIII)	(ull)	a1/a3
													• •		• •		•	• •	• •		
07/07	4	07/07	4	02:02	02:24	61	3.25	280		347	32	65	80	341	669	523	18	13			0.15
07/07	5	07/07	5	02:24	03:06	\$2	3.14	647		433	56	135	109	734	790	722	22	54			0.08
87/07	6	07/07	- 6	03:06	03:40	61	3.51	503		285	36	106	72	591	332	384	13	51			8.10
\$7/07	7	07/07	7	03:40	04:21	23	3.44	719		296	57	156	80	\$21	488	454	12	72			0.03
07/08	1	07/08	1	00:48	01:59	65	2.81	1365		2458	508	335	185	1055	3419	2547					0 05
07/08	2	07/08	2	01:59	02:25	57	2.87	798		1875	213	208	174	\$23	2446	1278					0 12
07/08	3	07/08	3	02:26	02:48	60	2.93	498		925	125	123	86	363	1557	1123					8 15
07/08	Å	07/08	Ā	02:48	63-10	61	3.02	467		683	180	111	115	355	1211	\$32					0 15
07/08	5	07/08	5	03-10	03-31	59	3.08	287		431	54	65	89	290	015	685					8 16
07/08		07/08	-	03-31	03-54	56	3 12	314		424	37	70	40	263	801	808					5.10
07/08	-	07/08	- 7	62-54	04 - 1R	60	3 64	274		428	45	£1	51	284	1844	687					6 14
77/08	.	07/09		84.14	04.10		3 61	214		-20		12		41	1000	143					0.14
87/09		07/08		04.10	04.57		3.31	33			64	13			114	163					9.25
61/00	10	07/00	10	04.13	94.54		9.31	803					£ a								8.00
01/00	10	01/08	14	94:32 	¥4:52	23	3.00	303			16		33	331	1200	831				•	
01/08	11	01/08	17	uninoi		26	2.80	662		801	190	ZUK	110	760	2142	1190					
														•••	• • •						
07/13	1	07/13	1	92:48	03:25	36	3.79	223		210	52	57	- 33	254	217	230	10	47	15	\$	9.05
07/13	Z	07/13	2	03:25	04:07		3.83	204		240	45	57	42	242	174	230			17	- 1	8.01
07/14	1	07/14	1	01:56	02:14	39	3.65	82		220	32	28	40	120	180	292	5		16	1	0.12
87/14	2	07/14	2	02:14	02:24	37	3.75	68		167	17	19	19	9 5	136	217					8.21
87/14	3	07/14	3	92:24	02:33	35	3.68	37		198	23	28	39	163	154	243			14	1	8.22
07/14	4	07/14	- 4	02:33	02:41	38	3.78	149		151	21	35	1	174	135	218					8.27
07/14	5	07/14	5	02:41	82:53	59	3.83	134		140	15	30	48	171	146	231	5		13	6	0.28
07/14	6	07/14	- 6	82:53	03:02	38	3.85	147		129	17	34	28	170	113	194					8.23
07/14	7	07/14	1	03:02	83:12	38	3.83	133		153	15	31	28	153	120	219	6		13	6	0.21
07/14	1	07/14		03:12	03:22	37	3.80	87		191	13	24	29	123	128	236					0.21
07/14	9	07/14	5	03:22	03:30	36	3.81	77		186	13	22	36	113	130	227	1		12	6	A. 25
07/14	10	07/14	10	03:30	03:38	44	3.91			148	16	26	25	132	112	182	•			•	8.31
07/14	11	07/14	11	03:38	03-47	36	3.82	97		154	13	25	32	135	127	364	1		14	6	8 77
07/14	12	07/14	12	03:47	03:56	39	3.81	#5		150	13	24	29	123	129	202	•		••.		8 74
87/14	13	07/14	13	03-56	04-07	45	1 79	65		150	13	19	10	160	1122	106			16	•	4 54
87/14	14	07/14	14	04-07	04-19	38	3 61	54		224	10	10	20	180	308	481				•	
87/14	15	07/34	15	04 - 18	84.33	40	1 14			557	14	27	50	124	- CV3 - CU7	691				••	0.10 0.10
87/14	16	07/14	16	64.22	04.45	28	3 37			454			67	115	685	893	61		10		8.10
67/14	17	87/14	17	M - 45	04.55	14	1 12	70		682	40	40 92	¥4 62	114	- 532 640	814	-			14	W. 10
07/14	1.	07/14	1.	32.40	05.00	26	3.36	70		748	42	23		1.44	343	810				14	9-21
07/14	10	07/14	10	NG - NG	05.17	11	3.33	86		497	€£	23	70	147	333	967			••		0.20
07/14	20	07/14	20		44.11 min	22	3.36	304		804	20	63	40	100	331	131			34	10	W. 13
67/14	20	01/14	20	CRAWLY .	ngin Volto	90	3.14	204		460	99 69	31	, 13	240	500	820	28		21	31	
017.4	41		41	TESC: 1		4	3.13	297		903	23	23	εų	233	221	Q 14					
01/14	•	87/14	•	18.05	18.64		3 96	184		-			**	121				••			
01/14		07/14		13:00	13:34	40	3.20	184		391			12	134	263	413	15	31	43	22	Q. 95
01/14	ć	07/14		13:26	20:21	56	3.23	182		391	44	41	120	206	541	521					0.08
97/14	3	01/14	3	20:21	20:39	37	3.41	147		313	33	36		151	364	413	10	\$3	35	13	0.11
01/14		07/14	4	20:39	20:54	39	3.47	132		254	23	32	43	137	305	355					0.15
07/14	5	97/14	5	20:54	21:11	40	3.48	121		Z15	22	29	41	125	251	386					0.13
07/14	5	07/14	6	Z1:11	21:26	41	3.50	122		194	18	28	33	152	243	283	•	62	20		0.15
07/14	1	07/14	1	21:26	21:39	39	3.53	96		182	15	25	41	123	221	272					8.17
07/14	t	07/14		21:39	21:51	39	3.54	140		197	22	32	37	138	245	295					Q.18
07/14	9	07/14	8	21:51	22:03	37	3.50	177		194	27	38	36	173	238	272	•	\$2	20	9	0.17
07/14	10	07/14	10	22:03	22:14	38	3.67	180		173	22	38	25	174	199	244					0.20
07/14	11	07/14	11	22:14	22:26	41	3.62	193		169	24	42	29	186	209	268					0.19

San Pedro Hill Cloudwater Data

Date	Seq	Date	Sec	Start	Stop	Vol	pH	Ne+	K+	MH4 +	Ca2+	Rg 2+	"Fo"	Ç1-	H03-	5042-	CH20	H202	HFo	NAc	LHC
						(∎])		(uli)	(UN)	(uii)	(uN)	(ull)	(ull)	(uli)	(uli)	(uli)	(ull)	(uiit)	(ull)	(ulli)	a]/ a 3
87/14	19	07/14	17	22.26	22.25	30	3 58	185		176	7.	41		107	435	282		53		-	
07/14	13	87/14	13	22.20	22.30	40	1 67	177		145	28	20	31 98	185	182	302	•	33	11	'	0.21
07/14	14	07/14	14	22:46	22:56	40	3.69	174		138	22	38	26	168	172	230					0.23
67/14	15	07/14	15	22:56	23:06	41	3.73	193		131	25	42	39	186	167	212	10	\$2	16	7	0.23
07/14	15	07/14	16	23:06	23:17	38	3.68	191		152	28	43	30	173	197	240					0.20
87/14	17	07/14	17	23:17	23:37	38	3.41	216		264	42	51	53	190	383	390					Ç.11
07/14	18	07/14	18	23:37	23:49	38	3.25	267		377	66	- 55	73	272	602	550	14	49	29	11	9.18
87/14	19	07/14	19	23:49	00:01	42	3.38	225		299	- 44	53	53	187	449	401					0.20
07/14	20A	07/14	20A	reserv	rin		3.44	52		205	17	18	25	57	325	284					
07/14	208	07/14	208	reser	voir		3.43	40		201	10	13	30	87	316	285	13	19	21	1	
87/15	,	07/15	,	13.18	13.38	18	2 88	342		764	180	10	147	315	867	1417					
87/15	3	07/15		19.45	26-50	54	3 28	274		754	199	76	130	181	865	1074					8.03 8 AE
07/15	ī	07/15	ī	20:50	21:18	12	3.23	172	۰.	745	158	£1	222	245	621	995					0.03
07/15	5	07/15	5	22:00	22:43	55	3.05	271		1141	236	100	248	357	1013	1583					0 07
07/15	\$	07/15	1	22:43	23:09	55	3.00	145		828	134	51	19	139	927	1214					8.12
07/15	1	07/15	1	23:09	23:33	56	3.07	89		538	81	34	81	117	674	876					0.13
07/15	1	07/15		23:33	23:56	55	3.08	79		482	68	30	\$3	120	685	805					8.13
07/15	9	07/15	9	23:56	00:55	55	2.64	198		2161	235	71	213	217	2797	2438					0.05
07/15	10	97/16	10	00:55	01:11	55	2.86	157		1225	165	58	154	218	1528	1438					0.19
07/15	11	07/16	11	01:11	01:25	56	3.04	101		606	\$0	36	108	158	880	872					0.22
07/16	12	07/16	12	01:25	01:38	56	3.14	65		427	55	24	78	112	640	643					0.24
07/16	13	07/16	13	01:38	01:53	59	3.12	55		507	52	22	82	-110	699	584					0.22
07/15	14	01/16	14	01:53	92:10	33	3.16	29		404	29	13	70	51	591	630					D.18
07/10	61 42	41/10	10	92:10	02:28	90 50	3.10			331		10	3/	40	347	54.8					0.17
07/10	19	07/16	19	Decan	92:44 Voie	20	3.21	20		330	91 14	12	**	41	544	438					9.19
	.,	9 17 10			VQ II		3.33	24		320		•	~~	• /	130	213					
07/16	2	07/15	2	15:35	16:07	55	3.56	#4		284			36	84	249	259					0.10
07/16	3	07/15	3	16:07	16:19	55	3.74	33		160			29	50	147	215					0.26
07/16	4	07/16	4	16:19	16:45	25	3.67	33		142			29	44	167	203					9.05
07/16	5	07/16	5	17:40	18:30	55	3.50	92		207			51	91	280	277					9.96
07/16	6	07/16		18:30	19:02	27	3.64	\$3		187	27	22	43	70	231	248					0.05
07/16	7	07/15		19:14	19:37	55	3.51	75		242	47	28	47	- #	285	329					8.14
01/10		01/10		19:37	19:49	96	3.55	32		126		••	18	22	145	235					8.25
07/16	10	07/10	10	20.03	20:03	20 56	3.80	33		34	13		13	33	110	170					8.10
07/16	11	07/18	11	20.00	20.33	55	4.00	24		90 90		,	13	30		614					8.51
07/16	12	07/16	12	20:33	20:50	56	4.15	20		88		,	10	27	17	104					11
07/15	13	07/16	13	20:50	21:04	56	4.30	19		56		i i		25	53	74					8.23
07/16	14	07/16	14	21:04	21:25	56	4.48	16		66	-	•	11	23	43	58					0.15
07/16	15	07/16	15	21:25	21:43	55	4.58	10		80				21	37	69					0.17
07/16	16	07/16	15	21:43	22:04	56	4.32	1		71				14	35	77					0.15
07/15	17	07/16	17	22:04	22:23	56	4.46	9		53				11	34	60					0.16
07/15	18	07/16	18	22:23	22:40	55	4.58	6		47	3	1		13	28	39					9.18
07/16	19	07/16	19	22:40	22:53	56	4.75	11		44	5	3		14	30	\$5					0.24
07/17	20	07/17	20	reserv	roir	33	3.80	49		504			33	79	300	376					
07/17	21	07/17	21	reser	voir	34	4.65	45		134			7	56	60	98					
				Number	-	240	242	241	63	24.2	225	231	236	242	267	24.2	80	16	21	21	227
				Kinim	u#-		2.42	5	3	44	3	1	1	11	28	39	5	4	12		0.00
				Maxim	un i		4.58	3795	307	\$383	1143	1151	586	2096	\$191	5796	51	72	43	31	0.34
				Arite	Ave		3 25	404	18	775	82	187	103	035	1196	817	99	45		10	0.14

Table 5 Henninger Flats Cloudwater Data

Henninger Flats Cloudwater Data

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Dete	Sec	Start	Stop	Vol	pH	Na+	MH4+	Ca2+	#g2+	¢1-	803-	S042-	CH20	H202	HFo	XAc	UIC
				(a))		(uN)	(uli)	(uli)	(UN)	(uN)	(ulit)	(uli)	(ull)	(ull)	(ull)	(ull) :	a1/m3
																-	
06/20	1	05:00	05:19	§ 5	3.59	272	1009	73	71	181	974	455	75				0.05
06 /20	2	06:15	06:56	45	3.35	180	805	39	41	169	877	447	75				0.07
05/20	3	06:56	07:36	24	3.19	192	\$21	46	52	177	1220	\$37	80				0.03
06/20	- 5	08:08	08:47	54	3.04	387	1461	90	103	308	3841	845	91				0.05
06/20	5	08:47	09:30	45	2.92	571	1743	117	155	452	2329	996	65				0.06
06/2C	1	09:30	11:03	43	2.75	1652	350\$	454	435	\$75	4034	2028	68				0.03
06/21	1	01:28	02:30	1	3.75	314	603	118	85	151	787	35					8.01
06/21	5	02:49	05:06	44	3.15	201	2411	249	257	496	2796	1371	76				9.02
06/21	6	05:05	05:53	43	3.04	454	1743	75	129	314	1982	1961	67				0.05
06/21	1	05:53	06:20	- 44	2.97	33G	1467	93	86	258	1802	\$34	65				8.05
05/21		86:20	06:34	45	3.04	243	1161	63	63	215	1420	173	- 54				8.18
06/21	9	06:34	06:50	44	3.05	193	\$45	- 45	- 54	202	1270	705	53				8.15
06/21	10	06:50	07:08	44	3.04	183	954	42	49	195	1252	704	58				8.14
06/2:	11	07:08	07:41	- 44	2.91	254	1229	\$0	67	223	1254	\$42	62				8.08
96/21	12	07:41	88:12	- 44	2.87	339	1487	85	91	255	1986	1094	- 64				9.08
05/21	13	08:12	08:39	44	2.14	434	1708	111	113	295	2233	1243	66				9.09
06/21	14	08:39	09:19	44	2.17	624	2122	188	157	400	3143	1542	75				8.06
86/21	15	09:19	10:03	18	2.62	1553	3172	520	400	665	5472	2606	105				0.02
07/09	1	23:47	01:38	36	3.88	127	1502	129	51	198	\$82	582					8.92
07/10	2	01:38	03:09	35	3.58	157	1760	167	63	214	1181	\$37					0.02
07/10	3	03:09	05:07	29	3.28	125	1530	127	52	197	1377	\$10					0.01
07/10	4	05:22	05:58	35	3.12	195	1295	159	99	200	1665	802					8.06
07/10	5	05:58	07:07	35	3.07	92	196	16	46	124	1327	438					0.03
07/10	6	07:07	07:31	35	3.21	50	795	47	20	101	\$23	368		-			0.08
07/10	7	07:31	08:08	34	3,16	55	143	49	20	111	193	412					8.05
07/12	. İ	08:08	08:31	36	3.15	65	\$54	61	24	112	1144	608					9 89
07/10		08:31	09:05	35	3.15	17	1103	91	30	117	1057	544					30.0
07/10	10	09-05	09.19	36	3 24	65	\$12	70	31	108		642					1 14
87/18	11	09-19	09.33	36	1 26	48	671	55	21		747	420					a 14
07/10	12	69-33	05.46	35	3 31	58	700	62	22		795	408					. 16
87/10	13	09.45	18.00	36	3 28	52	743		24		854	545					8 14
67/10	14	18.00	18-14	36	3 95	56	761		76	104	105	431					8 14
07/10	16	10.14	10.76	20	3.43	30	701	85		100	780	431					
67/10	16	10.96	10.20	36	3.20	30	880			180	100	69/ 69/					0.17
07/10	17	10.20	10.60	16	3.20	60	445	#1 #1	23 90	103	817	324					W. 11
01/10	- 10	10.60	10:00	30	3.20	33	012	102	23	149	115	601					0.14
07/10	10	11.16	11.30	33	3.13	13	837	102	31	121	1030	331					W. 12
07/30	13	11112)::30 	33	3.11		164	100	31	114	1132	1004					8.13
41716	20	(BPEL)	191E	03	4.80	34	1943	122	47	140	2000	1035					
07/1F	1	50.17	61.42	60	2 85		3507	146	34	22	1163						. 82
07/16		01.43	07.17		2 72	149	4085	234	75	69	1909	1474					B 67
37/16	í í	07.49	NS.EN	63	5 83	99	990J	204 196	13	50	1207	14/6					4.V.
07/16	-	02.50	02.50	42	5.03	E 8	3301	150		55	1911	800					0.00 0.05
07/10	2	04.50	05.94	84 67	5.09	30	3100	80	23	23	1211	300					0.05
01/10		100 D0	UJ:24	#2 #2	6.37 3 AF	4U 44	1947	1 H	13	43	9/9	112					¥.14
07/10		U3:24	00:43	83	3.93	33	1332	42	14	44	835	426					U.19
41/10		UD:43	UD:UD		5.12	25	1397	32	11	40	- 94]	404					8.14
01/16	8	90:00	UC:24	63	5.14	28	1172	31	11		1164	#42					1.22
07/15	10	46:Z4	06:35	H	3.46	50	774	43	17	41	683	400					0.30
07/16	11	06:36	06:45	- 54	3.65	50	597	40	17	38	478	310					8.40
07/16	12	05:45	06:55	- 14	3.65	41	571	31	14	- 54	464	294					0.36
07/16	13	06:55	07:05	\$4	3.69	31	562	23	11	59	482	315					9.36
07/16	14	07:C5	07:14	64	4.78	23	502	19	1	25	452	281					0.40
07/15	15	07:14	07:24	\$ 5	3.84	15	492	12		22	413	259					0.36
07/15	15	07:24	07:35	54	3.70	13	447	11	5	26	397	238					0.33
07/16	17	07:35	07:45	65	3.77	10	393	1	3	15	332	177					0.37

Table 5 Henninger Flats Cloudwater Data (continued)

Henninger Flats Cloudwater Data

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Date	Sec	Start	Stop	¥o] (≇1)	pH	Na+ (uli)	NH4+ (uN)	Ca2+ (uN)	Ng2+ (uN)	C1- (uN)	NC3- (uN)	S042- (uN)	CH20 (uff)	N202 (uff)	NFo (v≢)	HAc (ull)	LHC m]/m3	
07/15	18	07:45	07:54	54	3.80	6	324	7	3	. 18	284	139					0.40	
07/16	15	07:54	08:05	65	3.81	5	329	8	2	18	288	137					0.30	
07/16	2GA	reserv	voir	63	2.68	80	2118	397	77	115	1643	787						
07/16	208	reser	voin	31	2.83	52	1515	242	50	103	1164	54 8						
07/16	2	16:55	17:49	51	3.03	273	2435	\$8 5	171	301	3319	2028					0.07	
07/16	3	17:49	18:15	61	3.30	139	1235	518	\$6	155	1416	99 3					8.13	
07/16	- 4	18:15	18:40	62	3.29	85	1149	265	65	38	1167	796					8.15	
07/16	5	18:40	19:03	62	3.29	17	1016	234	59	20	1055	758					8.15	
07/16	6	19:03	19:28	\$3	3.20	- 64	1092	189	50	#1	1030	824					0.14	
07/16	· '¶	19:28	19:51	63	3.25	50	1020	136	41	- 80	1011	90£					0.15	
97/16	8	19:51	20:16	63	3.26	51	999	128	38	80	\$79	874					0.15	
\$7/15	9	20:14	20:35	63	3.26	50	\$48	125	36	76	\$29	741					0.17	
07/16	10	20:35	20:53	63	3.24	35	785	101	31	- 54	940	565					9.20	
07/16	11	20:53	21:11	54	3.22	31	741	87	28	48	1029	650					0.20	
07/16	12	21:17	21:28	64	3.27	23	\$39	56	20	39	\$27	447					9.21	
07/16	13	21:28	21:44	\$4	3.32	27	516	66	22	40	751	366					0.22	
07/15	- 14	21:44	22:00	65	3.41	18	398	43	16	22	504	274					0.23	
07/16	15	22:00	22:16	65	3.52	12	356	33	12	29	496	221					0.23	
07/16	15	22:15	22:34	65	3.48	-20	404	38	13	32	560	241					0.20	
07/16	17	22:34	22:55	- 64	3.49	- 17	387	32	12	32	513	212					0.17	
07/16	18	22:55	23:27	64	3.48	13	411	32	12	- 38	549	235					9.11	
07/17	19	reser	voir	40	3.94	3	273	15	7	27	231	155						
		N		76	76	76	76	75	76	76	76	76	17				72	
		Arith	. Avg.	3765	3.25	165	1203	118	57	142	1206	689	71				0.14	
		ffin	-		2.52	3	273	1	2	18	231	36	53				8.01	
		Max			4.78	1652	4085	985	435	\$75	5472	2606	109				0.40	

Date	940	SIARI	STOP	Tin+	3254+	Ca2+	H 2+	C1-	103-	5042-	114	EN 03
								neg /	1 3		· -	
06/10		01.00	05.00	170	110	96.1	44 3	e 0.2	147	113	-0 9	16 5
06/19	8	05.00	00.00	188	74	15 0	A2 A	105 4	81	124	-1 2	26.6
06/19	č	00.00	12.00	181	03	22.1	45.3	200.4	103	133	-1.2	27.1
06/19	ñ	20:00	00.00	115		24.8	33.5	51.5	102	104	14.5	28.4
06/20	Ā	02:00	06:00	60	73	17.2	29.5	60.2	94	93	2.9	10.7
06/20	B	08:00	12:00	168	89	17.8	45.2	72.3	99	127	-0.9	23.6
06/23	A	20:00	00:00	110	XA	19.9	26.2	34.1	187	294	-0.9	123.7
06/24	A	00:00	03:00	24	XA	14.4	2.5	7.9	51	37	-1.2	-0.5
06/24	В	03:00	06:00	67	XA	27.2	18.6	86.9	435	475	-1.2	12.9
06/24	C	07:30	08:00	13	318	16.1	6.7	0.0	43	340	-7.5	106.8
06/24	D	10:00	14:00	171	194	18.4	19.1	8.8	106	267	-0.9	140.5
06/24	E	14:00	18:00	173	162	34.9	48.8	39.5	147	188	38.4	131.8
06/24	F	20:00	00:00	59	447	17.5	26.0	7.6	211	317	-0.9	46.7
06/25	A	01:15	05:00	32	297	17.0	6.3	XA	32	338	0.6	17.3
06/25	В	05:00	09:00	5	17,	15.3	0.0	XA	7	22	19.1	-0.1
06/25	C	07:57	10:30	90	261	47.3	15.4	0.0	68	314	-1.5	162.7
06/25	D	11:00	15:00	99	197	30.4	26.3	0.0	81	255	6.8	133.0
06/25	2	16:00	20:00	XA	XA	AR	XA	, XA	NA.	39.	0.0	92.3
06/25	F	22:00	02:00	53	624	34.9	17.2	7.5	365	291	10.6	41.3
07/06		20:00	00:00	122	XA	23.8	26.4	64.4	189	131	-0.9	XA
07/07	A	00:00	04:00	70	MA	18.0	14.5	67.2	136	125	-0.9	21.1
07/07	6	09:00	12:00								0.0	33.9
07/07	5	09:00	12:00								49.2	36.3
07/07		14:00	10:00	- AA		10.0			20		0.0	20.7
07/12	.	20:00	02.00	12		19.9	3.0	0.0	20		10.0	-9.9
07/13		02.00	03:00			10.1	1.0			-	-1.4	70.0
07/13	2	03:00	12.00		976	90.9	1	20.1	30	100	104 8	180 0
07/13	ñ	12.00	15.00	76	120	92.3 98 6	17 1	2.1		102	41 6	100.2
07/13	ĩ	15.00	18.00	44	70	17 7	10.9	0.0		79	56.8	10.5
07/13	Ŧ	20.10	00.00	21	6R	27.4	11.5	0.0	20		28.8	22.0
07/14	i	02:00	06:00	14	161	6.8	1.7	0.0	6	180	-0.9	6.3
07/14	B	08:00	12:00	61	430	25.4	17.7	0.0	41	448	2.9	166.0
07/14	ē	14:00	18:00	73	MA	26.9	16.6	10.8	57	179	45.4	79.9
07/14	D	20:00	00:00	11	0	15.8	3.7	12.7	XA	42	-0.9	3.4
07/15	Ā	02:00	04:00	7	Ō	5.6	0.0	XA	23	32	-1.9	5.9
07/15	В	04:00	06:00	Ő	XĂ	0.0	0.0	MA		96	-1.9	0.9
07/15	Ċ	05:00	08:00	20	XA	. 7.1	0.9	19.9	124	166	-1.9	XA
07/15	D	08:00	10:00	7	KA	19.0	0.0	35.0	45	82	-1.9	MA.
07/15	E	10:00	11:30	0	XA	0.0	0.0	XA	16	75	-2.5	XA
07/15	2	14:00	18:00	24	NA.	17.8	7.3	16.3	36	101	19.9	XA
07/15	G	20:00	00:00	14)KA	38.8	8.3	26.8	137	200	-0.9	XA

NA indicates sample not analyzed

Tab]	le	7	Henninger	Flats	Aerosol	Data
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Date	Seq	START	STOP	Na+	NH4+	Ca2+	Ng2+	C1-	NO 3-	\$04 2-	MH3	20 103
							beg /		■3			
06/19		01:00	05:00	5	3	0.1	0.0	0.0	9	5	-0.9	12.8
06/19	В	06:00	09:00	0	5	0.0	0.0	0.0	8	5	-1.2	10.9
06/19	С	09:00	12:00	1	- 4	1.4	1.4	0.0	7	7	-1.2	17.5
06/24	A	02:00	06:00	33	77	24.3	11.8	2.3	51	74	2.9	65.4
06/24	в	06:00	10:00	36	117	12.5	13.9	0.0	83	102	-0.9	161.5
06/24	С	10:00	14:00	134	838	71.3	53.6	28.7	442	533	78.5	812.2
06/24	A	17:00	20:00	107	551	63.9	23.2	0.0	193	446	24.5	479.1
06/24	В	20:00	00:00	59	239	28.3	13.8	NA	114	189	30.7	47.4
06/25	B	14:00	18:00	106	645	92.9	27.3	22.5	287	479	98.6	897.0
06/25	С	20:00	00:00	38	284	32.8	9.4	0.0	83	234	18.4	78.4
06/25	A	02:00	06:00	54	250	34.7	14.8	NA	77	217	10.6	69.4
06/26	A	02:00	06:00	44	168	34.6	12.9	21.2	65	174	20.7	112.7
07/06	A	20:00	00:00	56	193	30.9	13.7	0.0	123	141	-0.9	63.1
07/07	A	02:00	06:00	29	75	16.4	7.6	0.0	46	66	-0.9	28.4
07/07	В	08:00	12:00	75	421	46.3	22.4	0.0	202	311	6.8	277.8
07/12	Å	20:00	00:00	21	93	33.5	9.3	7.8	28	81	26.8	72.9
07/13		02:00	06:00	- 4	27	2.9	4.0	0.0	12	33	9.1	17.4
07/13	B	08:00	12:00	21	183	27.9	13.7	0.0	99	96	91.7	251.2
07/14	A	20:00	00:00	35	151	27.7	13.2	NA	76	116	23.8	
07/15	Ă	02:00	06:00	15	111	10.7	4.0	0.0	20	95	1.4	
07/15	B	08:00	12:00	54	543	38.8	14.8	NA	254	313	58.5	

NA indicates sample not analyzed

Table 8 Kellogg Hill Aerosol Data

Date	Seq	START	STOP	Ka+	III 4+	Ca2+	Ng2+	C1-	1103-	8042-	113	1003
								neq /	B 3		•	
06/19		01:00	05:00	183	178	73.4	60.3	72.1	190	136	283.0	9.5
06/19	В	06:00	09:00	168	273	95.8	60.8	62.7	226	191	192.2	16.0
06/19	С	09:00	12:00	8	9	62.3	12.0	0.0	10	7	-1.2	6.1
06/24	A	02:00	06:00	140	581	64.6	41.9	15.6	452	291	-0.9	59.8
06/24	В	06:00	10:00	145	767	153.6	59.8	15.8	582	359	26.8	152.2
06/24	¢	10:00	13:50	149	875	192.8	61.6	0.0	567	451	131.9	624.0
06/24	D	20:00	00:00	124	250	78.3	35.8	0.0	234	203	34.6	52.7
06/24	E	02:00	06:00	125	594	109.3	25.6	0.0	392	372	26.8	63.1
06/25	B	20:00	00:00	84	279	51.3	28.2	10.9	191	167	58.5	52.3
06/25	С	02:00	06:00	62	549	40.1	11.1	12.0	372	271	34.6	43.4
06/25	D	08:00	12:00	66	838	116.3	32.2	0.2	496	441	154.9	385.8
06/25		08:00	12:00	115	784	179.0	48.8	XA	629	415	279.2	360.8
07/06		20:00	00:00	95	258	88.4	20.8	6.8	232	155	65.4	48.2
07/07		08:00	12:00	108	383	160.6	39.4	10.8	300	249	46.9	210.7
07/07		02:00	06:00	140	494	103.3	41.8	25.6	423	220	35.3	37.8
07/12		20:10	00:00	44	139	60.1	16.9	1.3	91	94	146.4	44.0
07/13		01:00	05:00	70	177	58.4	21.7	12.4	124	108	102.5	8.8
07/13	B	07:00	11:00	54	368	269.7	53.1	4.9	309	149	339.3	197.4

NA indicates sample not analyzed

Table 9 Collector Blank Analyses

Dete	Site	Baseription	Ke+	1014+	Ca2+	Mg2+	¢1-	W03-	S04 2-
			(uli)	(ull)	(ulli)	(ulii)	(ull)	(uiii)	(uli)
35/13	SPH	reservoir only after rinse	٥	8	٥	٥	٥	0	đ
06/13	SPH	first after rinse	2	i	3	1	1	ŝ	0
36/13	Spu	second after rinse	1	3	1	0	- 6	0	C
05/18	SPn	reservoir only after rinse	7	2		2	13		9
36/18	SPH	blank #1 after rinsing	5C	16		10	49	22	21
06/16	SPH	blank \$2 after minsing	27	10			32	17	18
06/20	SPH	clear bottle left in fridge	0	8	9	0	- 6	0	9
06/21	SPH	blank after rinsing	78	16	- 14	20	- 58	48	26
06/22	SPH	blank after rinsing	40	- 54	13	12	33	77	71
06/23	SPH	reservoir only after rinse	5	1	1	2	1	15	11
06/24	SPh			- 64	4		6	44	52
06/25	SPH	blank after cleaning			5		23	80	74
87/02	SPH	blank after rinsing.	21)	83	39	54	134	228	144
07/06	SPh	blens after rinsing		30	4	5	20	47	46
97/07	SPH	unused bottle		0	0	0	0	0	9
67/67	SPH	blank after minsing		11	3	5	25	20	15
07/12	SPH	blank after rinsing	2	3	4.	2	5	1	6
07/13	SPH	blank after rinsing	3	1	1	2		12	
07/13	SPH	blenk at 2000	- 24	17	10		- 16	33	29
07/14	SPH	blank after rinsing	15	27	ļ	5	3	22	. 42
07/14	SPH	blank after rinsing	5	11	4	2	1	- 11	15
0//15	341	blank on large roos	8	Q	Q	Q	g.	9	I
06/09	HF	blank after rinsing -	1	2	8	9	8	8	
06/05	HF	blank after rinsing	1	3	0	0		0	. 0
06/14	HF	blank after rinsing	1	27	7	3	7	0	32
06/15	HF	blank after rinsing	5	6	2	1	0	0	- 0
96/18	HF	rinse blenk	3	15	2	1	11	20	11
86/15	NF	sitting overnight w/o rinse	57	80	30	19	18	133	72
05/19	NF	after 100 ml rinse	18	27	10	5	10	- 44	24
86/20	HF	reservoir only after rinse	13	25	1	3		31	20
06/20	MF	system after rinse	33	50			14	75	36
96/22	MP MC	post cleaning strands, left in tridge	5	19	2	2	3	23	15
00/23	HP NC	Blank atter rinsing		- 21			21	48	23
00/24	HE	plank atter rinsing			ų,			U A	
06/23	nr Mt	TESETVOIT ONLY STORE FINSE	1						
06/30	WE	bienk ei eiler rinsing bissi 97 sfear singing		17	:		-		
07/02	HE	blank after singing		19				10	
07/02	NE	bland after cinetan	,		,		() ()	15	14
27/07	MC.	blank after Pinsing blank after Pinsing			11			12	
87/88	HF	black after ringing	147	314	1.	•		410	205
07/05	NF	personal ofter since		0	ė	n			200
07/09	HF	everas after mines	10	1	;		i	12	
07/05	HF	EVELOF After rinse	6	3	1	ć	;	10	6
07/10	HF	blank after rinsing		1	,	ă	,		5
07/*:	HF	rinse of bottle sitting in fridge	۵	č	ċ	0	21	Č.	2
07/12	HF	before rinsing, sat 2 days w/o sampling	19	117	39	15	10	182	10
67/12	HF	after ransing	1		;	0		6	
\$7/13	HF	collector only, left in fridge overnight	1	,	3	1		14	Ē
07/14	H.	blank after rinsing	3	11	6	1	5	18	Ì
07/14	HF	blank after minsing	3	11	6	2	i	17	10
07/15	HF	blank after rinsing	2	104	2	1	6	17	C
07/15	HF	blank after rinsing	•	60	2	0	1	21	0
67/17	HF	black after rinsing	t	10	•	0		11	ĉ

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Table 10 Size-fractionated sample concentrations

Date	f/b	Seq	Start	Stop	Vo]	pH	Na+	NH4+	Ca2+	Mg2+	¢1-	803-	S042-	S(IV)	CH20	H202	NFo	NAC	-/+	Rate
					(#1)		(uli)	(uli)	(IN)	(uN)	(UN)	#)	(ull)	(uff)	(ull)	(ull)	(ull)	(ull)	-	al/sin
07/15	Ŧ	91	84 - 86	64 - 30	70	3 40		100	,	£		164	96 1							• •
67/15	÷	22	04.33	05-00	RR .	1 24	10	251	12		24 23	330 185	155		18	10	18		1.04	2.3
07/15	÷	23	05 - 00	05.30	71	3 27	48	171		12	75	224	461		16			11	1.00	2.3
07/15	, ¢	24	05:30	86.00	71	3 25	52	615	65	16	70	£18	585		27		401 411	10	1.00	2.4
87/15	f	25	06-00	06.30	74	3 29	55	222	40	19	23	706	777		50	Ň	**	1.8	0.00	2.4
37/15	÷	26	06:30	07:00	112	3.42	87	391	22	21	£1	300	136		15		26	51	1 82	2.3
07/15		27	07:00	07:30	121	3 42	55	202	12	14	12	276	242		13		17		1.02	3.7
07/15	ŕ	28	07:30	08:00	136	3.44	35	191		10	58	218	128		19	i	10		0.00	4.5
07/15	÷	29	08:00	08:30	122	3.49	25	192		1	42	222	248		12	10	10	10	1 02	•
07/15	- ÷	30	08:30	89.00	116	3.52	23	166		÷.	40	204	271		13	16	90		1 82	1.4
07/15	ŕ	31	09:00	05:30	114	3.56	45	164	17	13	4	188	24.7					•	8 83	3.3
07/15	÷	32	09:30	10.00	110	3 64	30	138	13	11	122	178	214						1 10	3.8
87/15	ŕ	33	10-00	16-30	56	3 42	27	234	20	10	15	187	278							4.1
87/15	÷	34	10:30	11:00	- 64	3.40	41	275	54	19		117	418		18	44	22	12	1 44	3.3
07/15	4	35	11-30	12.00	40	3 54	161	274	217	57	116	141	664			43	**	13	1.89	2.1
07/15	÷.	36	12-00	13-00	30	3.38	478	432	\$15	175	\$25	710	1087						1.02	6.5
•••	•	••				3.30		455	313										1.99	8.3
			Sampl	es froi	. *** *	stran	ds: Se	mail dr	opleta	;								·		
07/15	Ь	21	94:00	04:30	43	3.55	,	171	4	,	25	233	192							14
07/15	Ь	22	04:33	05:00	29	3.42	11	267	5	. 3	57	326	361	•	11	•	16	18	1 63	1 1
07/15	ь	23	05:00	05:30	21	3.78	12	376	Ē	Ĩ	57	478	477		19		18	11	1.65	
07/15		24	05:30	06:00	31	3.10	13	804		1	100	123	684				- 14	17		• 1 A
07/15		25	06:00	06:30	28	3.08	14	1479		i	118	1152	1016		23	- 1		99	8 88	
07/15		26	06:30	87:00	38	3.24	11	445	Å	,		728	692		25		17	50	A 44	1.2
07/15	6	27	07:00	07:30	39	3.35	13	424		i	76	401	432		17		18		1 82	1.3
87/15	b	28	07:30	08:00	38	3.38	12	111	3	3	52	303	141		16		18		1.94	1.3
87/15	ъ	29	08:00	08:30	30	3.42	16	317	,	Š	51	281	197		17		10	18	1 81	1.3
07/15	- b	30	08:30	09.00	32	3 47	18	260		7	52	747	152		14			14	1 85	1.0
07/15	ь	31	09-00	09-30	22	3.49	10	242	Š		- u	187	980			••	**		8.86	
07/15	ь	32	09:30	10:00	35	3.59	14	205	,	5	105	184	250						1 11	1 2
07/15	b	33	10:00	10:30	16	3.45	21	282	15	i	50	256	350						8 87	1.2
87/15	- b	34	10:30	11:00	30	3.25	35	488	22	14	123	457	606		21	11	40	17	1.46	1.6
87/15	b	35	11:30	12:00	16	3.74	29	478	30	12	46	408	654		• •	+*	••	••	1 81	1.5
07/15	b	36	12:00	13:00	14	2.96	76	685	73	30	172	705	1143						1.03	0.2
		- .																		"LHC"
		Comb	ined V	lues																(8)/83
07/15		21	04:00	04 : 30	112	3.45	14	182	6	4	33	309	229						1.02	0.21
07/15		22	04:33	05:00	\$7	3.36	24	256	10	5	63	376	339		10				1.05	0.20
07/15		23	05:00	05:30	99	3.27	38	338	18	10	70	469	453		15				1.05	0.18
\$7/15		- 24	05:30	06:00	102	3.20	41	\$72	41	12	79	680	622		27				0.99	0.19
07/15		25	06:00	96:30	101	3.22	50	1057	31	15	79	\$27	809		36				0.98	0.19
07/15		26	06:30	07:00	150	3.37	67	532	18	16	\$3	483	501		18				1.00	0.28
07/15		27	07:00	07:30	150	3.40	45	256	10	12	73	307	365		14				1.04	0.30
07/15		28	07:30	98:00	174	3.43	31	224	7		57	238	342		14				0.99	0.32
07/15		29	08:00	08:30	151	3.48	23	216	6	1	44	234	318		13				1.01	8.28
07/15		30	08:30	09:00	147	3.51	22	186	8	1	42	212	288		13				1.82	0.28
07/15		31	09:00	09:30	136	3.55	39	177	15	11	46	188	248						8.82	0.25
07/15		32	09:30	10:00	145	3.63	33	154	12	10	118	120	223						1.17	0.27
07/15		33	10:00	10:30	71	3.43	26	244	19	10	38	206	255						8.74	8.13
07/15		34	10:30	11:00	- 54	3.35	44	344	- 44	17	97	377	478		19				1.85	0.18
07/15		35	11:30	12:00	57	3.43	123	332	164	44	110	360	590						1.03	0.11
07/15		36	12:00	13:00	44	3.20	351	\$12	374	129	277	722	1105						1.85	0.04



Figure 1. Map of the Los Angeles area showing the seven sites used for sampling cloud and fogwater, aerosol, and selected gases during the summer and winter portions of the South Coast Air Quality Study (SCAQS). Sites are depicted by a bold dot. Also shown on the map are major freeways and mountains.







Figure 3. pH distribution of intercepted stratus cloudwater samples collected at Henninger Flats during the summer of 1987.



Figure 4. Time trace indicating rapid changes of observed species concentrations, pH, and liquid water content during the morning of June 25, 1987, at San Pedro Hill. Samples were collected as rapidly as one every ten minutes by the CASC and the autosampler during this period.







Figure 5. Performance of the fractionating collector is illustrated for three different liquid water contents. Shown for each liquid water content are the initial droplet distribution (based on Best's formula; Best, 1951), the portion of the droplet spectrum collected on the large rods in the fractionating inlet, and the portion collected by the CASC strands in the main body of the collector. The collection efficiencies are based on impaction theory and the percentage of airflow through the collector sampled by each collection surface.



Figure 6. Comparison of sample volumes, Na⁺ concentrations, and Mg^{2+} concentrations for the samples collected by the fractionating collector on the morning of July 15, 1987. Samples collected on the large rods in the sampler inlet are labeled "large drops", while those collected by the CASC strands are labeled "small drops."



Figure 7. Comparison of Ca^{2+} concentrations, H⁺ concentrations, and Cl⁻ concentrations for the samples collected by the fractionating collector on the morning of July 15, 1987. Samples collected on the large rods in the sampler inlet are labeled "large drops", while those collected by the CASC strands are labeled "small drops."


Figure 8. Comparison of NO_3^- concentrations, SO_4^{2-} concentrations, and NH_4^+ concentrations for the samples collected by the fractionating collector on the morning of July 15, 1987. Samples collected on the large rods in the sampler inlet are labeled "large drops", while those collected by the CASC strands are labeled "small drops."



Small Droplets



Combined Sample



Figure 9. Illustration of the composition of the cloudwater sampled between 0600 and 0630 on July 15, 1987, using the fractionating collector. The fraction labeled "large droplets" was collected on the large rods in the inlet. The fraction labeled "small droplets" was collected on the CASC strands in the main body of the collector. The "combined sample" represents the volume weighted average composition of the two fractions. The area of each diagram is proportional to the total of the measured ionic species concentrations in that fraction of the sample.

CHAPTER 4

CHEMICAL COMPOSITION OF COASTAL STRATUS CLOUDS:

Dependence on Droplet Size and Distance from the Coast

by

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Abstract

The aerosol at elevated sites in the South Coast air basin is a mixture of sea salt and pollution-derived secondary aerosol. The influence of sea salt declines with increasing distance from the coast. Nitric acid appears to react with the NaCl in sea-salt aerosol to release $HCl_{(g)}$ and form NaNO₃ in the aerosol. At inland sites aerosol concentrations differ during onshore and offshore flow. The highest concentrations are observed during the day when the onshore flow transports pollutants to the sites, while lower concentrations were observed at night when drainage flows from nearby mountains influenced the sites. Variations in liquid water content are a major influence on cloudwater concentration.

Comparisons of the ionic concentrations in two size-segregated fractions collected during each sampling interval suggest that there is a large difference between the average composition of the smaller droplets and that of the larger droplets. For each time interval, the concentration of Na⁺, Ca²⁺ and Mg²⁺ in the large droplet fraction was observed to be higher than in the small droplet fraction. The concentrations of SO_4^{2-} , NO_3^- , NH_4^+ , and H⁺ were higher in the small droplet fraction. Chloride concentrations were nearly equal in both fractions. Differences in the composition of size-fractionated cloudwater samples suggest that large droplets are formed from sea salt and soil dust, which are large aerosol, and small droplets are formed on small secondary aerosol composed of primarily ammonium sulfate and ammonium nitrate. The concentrations of several components that exist partly in the gas phase (e.g. Cl⁻, HCOOH, and CH₃COOH) appear to be independent of droplet size.

Introduction

Coastal stratus clouds are a major feature of the Los Angeles weather pattern. Cass (1979) noted a correlation between episodes of high SO_4^{2-} concentrations and the presence of cloud or fog. Previous studies by Waldman et al. (1985) and Richards et al. (1983) have indicated that stratus clouds above Los Angeles have high concentrations of NO_3^- and SO_4^{2-} .

The size-dependent composition of aerosol is well known. Soil dust and sea salt generally make up the large aerosol, while nitrate and sulfate, partially neutralized by ammonium, make up much of the sub-micron aerosol. If droplet size is dependent on the size of the condensation nucleus then the composition of droplets could depend on size as well. In Japan, Naruse and Maruyama (1971) observed a correlation between droplet size and nucleus mass. The larger droplets contained large sea-salt nuclei, while the smaller drops contained ammonium sulfate Hudson and Rogers (1984) indirectly determined nucleus size by measuring the aerosol. critical supersaturation of nuclei from different size droplets. They found that the largest drops contained nuclei with low SS_{cr} (i.e. large mass). Noone et al. (1988) have recently provided evidence that solute concentration within cloud droplets can be a function of droplet size in coastal stratus clouds (Cheeka Peak Research Station, Washington). They reported that cloud droplets in the size range of 18 to 23 μ m had solute mass 2.7 times that of droplets in the size range of 9 to 18 μ m. Their results were consistent with the suggestion by Andreae et al. (1986) that in marine air the larger droplets are nucleated on dust or sea salt, while the smaller droplets form on excess (non-sea-salt) sulfate derived from the photoxidation of dimethyl sulfide. Collett et al. (1988) attributed differences in the composition of cloudwater samples collected by two collectors with different 50% lower droplet size cuts to size-dependent differences in droplet composition. Some of the theoretical aspects of size-dependent variations in droplet composition have been discussed by Perdue and Beck (1988).

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In addition to an anticipated dependence of the chemical composition of the coastal stratus on droplet size, we also expected to see significant variations in composition as a function of distance from the coast. In order to acheive these objectives we developed a cloudwater sampler capable of size fractionation and we established cloudwater and aerosol sampling stations at three elevated sites in the Los Angeles air basin during a period of prolonged cloud cover. This effort overlapped in time with the Southern California Air Quality Study (SCAQS) of 1987.

Methods

• Site Descriptions

Three sites, San Pedro Hill, Henninger Flats, and Kellogg Hill (see Fig. 1), were operational during the period of June 13 to July 17, 1987. The San Pedro Hill site (elev. 450 m) was located at a radar and communications facility operated by the U. S. Air Force and the Federal Aviation Administration. San Pedro Hill is the easternmost hill of ridge that forms the Palos Verdes Peninsula. The distance from the site to the ocean was 2.5 km; Los Angeles Harbor was 6 km east. The sampling equipment was placed at the edge of a flat grassy area. A steep hillside sloped away from the site, giving it unobstructed exposure from $70-270^{\circ}$.

Henninger Flats (elev. 780 m) is on a level bench partway up Mount Wilson in the San Gabriel Mountains, 7 km NE of Pasadena, and 45 km NE of the coastline. This site has been used in previous sampling programs (Waldman et al., 1985) and for a cloud/fog collector intercomparison (Hering et al., 1987). The Flats is an experimental forest area that has been planted with pine trees; the sampling equipment was located in a clearing a few hundred meters back from the edge. Because it is partially sheltered by the surrounding trees, cloud interception at the samplers is slightly delayed.

Kellogg Hill is 38 km E of downtown Los Angeles at an elevation of 370 m. It lies 50 km NE of the coastline. The sampling equipment was located in a fenced enclosure adjacent to a small building that housed radio transmission equipment. The building partially obstructed

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the sampler when winds were from the south to west, which is the prevailing daytime wind. The site was unobstructed in the direction of the prevailing night-time winds. Construction activity near the site increased aerosol concentrations of soil dust during the daytime.

• Sampling Procedure

Each site was equipped with a Caltech Active Strand Collector (CASC) with an automated fractionating sampler and a cloudwater sensor, shown in Figure 2 (Daube et al., 1987). The CASC collects droplets by inertial impaction on 510 μ m Teflon strands. The 50% collection efficiency cutoff, predicted from impaction theory and based on droplet diameter, is 3.5 μ m (Friedlander, 1977). A protective rain shield, which had its opening facing downward, was attached to the front of the collector to exclude large (d > 200 μ m) sedimenting droplets. The cloudwater sensor, which is a miniature version of the CASC connected to a resistance grid, was used to turn the collector on when fog was present. The cloudwater collected by the CASC was directed to the fractionating sampler. In the fractionating sampler, cloudwater accumulated in a reservoir until a liquid level sensor determined that 60 ml had been collected. The reservoir was then drained to a 60 ml polyethylene sample bottle held in a carousel. The carousel had a capacity of 20 bottles. When all 20 bottles had been filled, the level sensor was deactivated. Any further sample collected was retained in the reservoir. If its capacity was exceeded the sample drained out through an overflow tube. The reservoir and sample bottles were housed in a refrigerator. A printer recorded the times that the sampler came on and off and when the bottles were filled. Also located at each site was an automated filter pack aerosol sampler. Open-faced Teflon filters (Gelman Zefluor, $1\mu m$ pore size) were used to collect aerosol for inorganic analysis. $HNO_{3(g)}$ was collected on a nylon filter (Gelman Nylasorb) placed behind one Teflon filter. $NH_{3(g)}$ was collected on two oxalic acid impregnated glass fiber filters behind a second Teflon filter. A rain shield above the filter holders excluded debris and sedimenting droplets.

During the late afternoon or evening prior to an expected cloud event, the samplers were cleaned by rinsing the collection strands, sample tubing, and reservoir with distilled, deionized

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water (DDH₂O). After rinsing, the strands were sprayed again with DDH₂O, which was collected in the fraction collector as a system blank. Rinsing and blank collection were repeated the following morning whether cloud was collected or not. Three sets of filters were loaded on the aerosol collector. A timer on the collector controlled the times that each filter set was run.

At San Pedro Hill, a size-fractionating inlet was used on the CASC for the period of 15–16 July, 1987. The rain shield was not used in conjunction with the size-fractionating inlet. Four rows of eight 12.7 mm Teflon rods, which have a 50% lower size cut of 16 μ m (diameter) at the sampling velocity of 9 m s⁻¹ are arranged at the front of the inlet. Each row covers 46% of the cross-sectional area. Large droplets impact on the rods, while most droplets smaller than 15 μ m pass through to be collected on the CASC strands in the main body of the collector. Water from the rods and strands is collected in separate bottles, which were manually emptied at the end of each sampling interval.

Analytical Procedures

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Samples were retrieved in the morning following a cloud event and transported to the lab at Caltech. The samples were weighed to determine their volume and the sample pH was measured with a Radiometer PHM82 pH meter using a combination electrode calibrated against pH 4 and 7 buffers. For selected samples small aliquots were removed and treated to stabilize reactive species. CH_2O was stabilized by addition of SO_3^{2-} (Dong and Dasgupta, 1987). Likewise, in a separate aliquot S(IV) was stabilized with CH_2O . Hydrogen peroxide was reacted with p-hydroxyphenylacetic acid in the presence of peroxidase to form a stable dimer (Lazrus et al., 1985). Carboxylic acids were preserved by addition of chloroform (Keene and Galloway, 1984).

The samples and preserved aliquots were stored in a refrigerator at 4 $^{\circ}$ C until analysis. Major anions were determined by ion chromatography with a Dionex AS4 or AS4A separator column and a micromembrane suppressor. The eluent was 2.8 mM HCO₃^{-/} 2.2 mM CO₃²⁻. The metallic cations were determined by atomic absorption spectrophotometry. An

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air/acetylene flame was used for Na⁺ and K⁺; N₂O/acetylene was used for Ca²⁺ and Mg²⁺ to minimize interferences. NH_4^+ was determined by flow injection analysis employing the indophenol blue method.

The stabilized CH_2O was determined by a modification of the Nash method for use with an autoanalyzer (Dong and Dasgupta, 1987) Hydrogen peroxide was added to eliminate S(IV), which interferes by forming an adduct with CH_2O . The absorbance of the colored product was measured at 412 nm. S(IV) was analyzed by the pararosaniline method (Dasgupta et al., 1980) adapted for flow injection analysis. Carboxylic acids were determined by ion exclusion chromatography (Dionex ICE-AS1) with dilute HCl as the eluent and by normal ion chromatography using $Na_2B_4O_7$ as the eluent.

The Teflon and oxalic acid-impregnated glass fiber filters were extracted in distilled deionized water (DDH₂O) on a shaker table. A small volume of ethanol was added to the filter prior to extraction to more effectively wet the filter surface. The nylon filters were extracted in HCO_3^{-}/CO_3^{2-} IC eluent. Composition of the extracts was determined by the same procedures used for the fogwater samples, with the exception of additional buffer in the complexing reagent and oxalic acid in the rinse solution of the ammonia analysis to account for the effect of the oxalic acid. Complete details of the analytical procedures and estimates of their precision and accuracy are presented elsewhere (Munger, 1989).

Results

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Aerosol Composition

The aerosol and gas-phase data for San Pedro Hill, Henninger Flats, and Kellogg Hill are summarized in Table 1. The samples are separated into day and night samples to account for the diurnal variation in wind direction at the sites. Onshore winds prevailed during the day; offshore drainage flows prevailed at night. The major species present were NH_4^+ , NO_3^- , and SO_4^{2-} . In many of the daytime samples HNO_3 was equal to or greater than the NO_3^- . In general, the concentrations of N(-III), N(V), and SO_4^{2-} increased with distance from the coast.

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Na⁺, Cl⁻ and Mg²⁺ decreased away from the coast. The highest concentrations of NH_3 were observed at Kellogg Hill.

Day and nighttime concentrations at San Pedro Hill were similar. At Henninger Flats and Kellogg Hill there was a tendency towards higher concentrations in the daytime samples. This apparent trend was not statistically significant, however, because of the large degree of variability in the daytime concentrations.

Most of the samples showed a large Cl⁻ deficit when compared to the sea salt Na⁺:Cl⁻ ratio (see Figure 3A). The averages in Table 1 indicate that the Cl⁻ deficit is generally greater in the daytime.

Concentrations of the acidic anions, $NO_3^- + SO_4^{2-}$, exceeded those of NH_4^+ in all but one sample from San Pedro Hill (Figure 4A). The overall acid-base balance of the atmosphere is indicated in Figure 4B which plots total acids against total bases (acids = $Cl^- + NO_3^- + SO_4^{2-}$ + HNO₃; bases = $NH_4^+ + Na^+ + Ca^{2+} + Mg^{2+} + NH_3$). The difference between the two sums is equivalent to total atmospheric alkalinity, as defined by Jacob et al. (1986). Most of the data plot along the 1:1 line, which indicates neutrality. The samples with excess acids are from Henninger Flats and Kellogg Hill. Kellogg Hill also had the only samples with excess bases.

Cloudwater Composition

The weather pattern during June and July 1987 was ideal for cloudwater collection. Clouds formed nearly every night. During the sampling period, 242 samples from 18 cloud events were collected at San Pedro Hill. Because the sampling carousel could only hold 20 time-resolved samples, long cloud events with high liquid water content (LWC) were not sampled to the end.

Concentration data for the San Pedro Hill cloudwater samples are summarized in Table 2. Typical LWC values, as estimated from collection rate and the theoretical collection efficiency of the CASC, were $\simeq 0.1$ g m⁻³. Cloudwater at San Pedro Hill was consistently acidic. The overall range of pH was 2.4 - 5.0; the volume-weighted average pH was 3.15. The major

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anions were NO₃⁻ and SO₄²⁻; on an equivalent basis NO₃⁻ was in slight excess. In addition to H⁺, the major cations were NH₄⁺ and Na⁺. Their ranking varied from event to event. Concentrations of CH₂O were typically 20 - 30 μ M, while the concentrations of formate and acetate were found to be in the range of 12 - 43 μ M and 6 - 31 μ M, respectively. Hydrogen peroxide concentrations were found in the range of 4 - 72 μ M while S(IV) was usually absent in samples for which it was determined. In addition to formaldehyde other aldehydes such as acetaldehyde (1 - 5 μ M), glyoxal (1 - 10 μ M), and methylglyoxal (4 - 8 μ M) were identified and quantified in cloudwater collected at San Pedro Hill during this sampling program (Igawa et al., 1988).

Clouds intercepted Henninger Flats less frequently than at San Pedro Hill. Because of the difference in their elevations, clouds did not usually intercept both sites simultaneously. At Henninger Flats, 76 samples from 5 cloud events were obtained during the sampling period. Cloudwater from Henninger Flats was similar in chemical composition to that from San Pedro Hill (see Table 4). The range of pH was comparable at the two sites, but Henninger Flats had a slightly higher pH on the average. Henninger Flats had higher concentrations of NH_4^+ and lower SO_4^{2-} and Na^+ . Because of the lower SO_4^{2-} , the ratio of NO_3^- to SO_4^{2-} was greater at Henninger Flats. Stratus clouds did not penetrate far enough inland at low elevation to permit sampling at Kellogg Hill.

In the majority of the samples, Cl⁻ and Na⁺ were present at the sea water ratio (see Figure 3B). A Cl⁻ deficit (or Na⁺ excess) was consistently observed in samples with Na⁺ > 750 μ N. The apparent Cl⁻ deficit in the average concentrations is due to the influence of the high concentration samples.

• Temporal Variations

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Concentration vs. time profiles for 4 representative periods are shown in Figures 5 - 8. Gaps in the LWC trace indicate non-continuous sampling when the 20 bottles had been filled. Concentrations for the period 0400 -1300 on July 15 at San Pedro Hill are derived from the volume-weighted average of the two size-fractioned samples. Concentrations tended to

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decrease as LWC increased, and then increase again at the end of the event as LWC decreased. Some depletion was also apparent during long periods of stable LWC, such as the samples after 0700 on July 15 at San Pedro Hill and the second set of samples from Henninger Flats on July 16.

Most of the samples obtained at the two sites were collected during non-overlapping time periods. Often the stratus clouds would intercept one site on a given day and not the other. At other times, the clouds were observed to intercept the hillside at San Pedro during the period shortly after midnight, while interception at Henninger Flats did not begin until a few hours later. By this time, clouds were no longer intercepting the slopes of San Pedro Hill. The only event with simultaneous collection at both sites was on the night of July 16-17. This event, which was associated with drizzle in the L.A. Basin, was one of the most persistent during the study. Clouds were present at both sites the previous night, though the bottles at San Pedro Hill were full by 0244 on 16 July when continuous cloud interception began at Henninger Flats. The clouds did not completely clear during the day of July 16. Continuous cloud cover returned to both sites around 1740 the evening of July 16 and filled all 20 bottles by approximately 2300 on 16 July. The clouds continued without interruption until 0500 at both sites and was intermittent until mid-morning. This event had the lowest overall concentrations of major ions and the highest pH observed at San Pedro Hill. The pH and ion concentrations at Henninger Flats were close to their average values.

Size-fractionated Samples

The size-fractionated cloud samples were collected during an extended cloud event at San Pedro Hill. Clouds initially intercepted the site at approximately 1900 on July 14 and remained until approximately 1300 on July 15. Twenty samples were collected between 1900 and 0000. Collection of size-fractionated samples began at 0400 and continued until 1200. A final one-hour unfractionated sample was collected from 1200 to 1300.

The overall concentration in the clouds when the size-fractionated samples were collected

is indicated in Figure 6. The major ions are comparable to those at the beginning of the event and in the subsequent event. The sea salt ions were less than in the preceding and following samples. Figures 9 - 12 illustrate the major ions, sea salts and collection rates in the two fractions. The concentrations of all species increased concurrently around 0600 without appreciable change in collection rate. The sharp increase in the collection rate for large drops subsequent to this corresponded to a period of drizzle. Concentrations gradually decreased over the next several hours until the final samples when they rose again as LWC dropped.

Discussion

• Inter-site and Temporal Variations

The differences in chemical composition at the three sites are consistent with their location. San Pedro Hill, being the closest to the ocean, is the most affected by sea salt. During the day, prevailing winds carry emissions of NO_x and SO_2 and their oxidation products, HNO_3 and SO_4^{2-} , inland where they impact Henninger Flats and Kellogg Hill. Higher N(-III) levels are obtained near the dairy feedlots in Chino (see Figure 1). As noted above, the ratio of SO_4^{2-} to NO_3^{-} in cloudwater is higher at San Pedro Hill than at Henninger Flats. Local oil refinery emissions, which affect San Pedro Hill at night, when offshore winds prevail, may directly influence this ratio.

Aerosol concentrations at Henninger Flats and Kellogg Hill were generally less at night than during the day. Henninger Flats was frequently above the inversion at night, which could explain the low concentrations there. In addition, downslope flow from the adjacent mountains, which rise to 1800 m, could flush the site with cleaner air at night. Kellogg Hill, which is also near the San Gabriel Mountains, may also be affected by this mechanism. San Pedro Hill, on the other hand, is far from any higher ground and will not be affected by clean-air drainage flows. The offshore winds that influence the site at night may have been influenced by primary emissions and secondary pollutants from the SCAB. A sea breeze during the day would normally be expected to bring relatively clean air to the San Pedro Hill

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site. However, as Cass and Shair (1984) have pointed out, pollutants accumulate off the coast of Los Angeles due to the daily sea breeze/land breeze cycle. Thus, the onshore flow of the sea breeze returns pollutants that were carried out to sea by the previous nights offshore flow.

Acid-Base Balance

The routine observation of pH 3 cloudwater clearly indicates that the airmass in the portion of the SCAB affected by stratus has excess acidity. The acid-base balance at San Pedro Hill and Henninger Flats (Fig. 4B) is generally consistent with this observation. Previous observations by Jacob et al. (1985) noted that coastal areas are generally deficient in acid-neutralizing capacity. Further inland, near the major sources of NH_3 (e.g. cattle feed lots and other agricultural activities), samples with excess alkalinity were collected. In the presence of excess acidity, sea salt aerosol readily loses $HCl_{(g)}$ if the humidity is <99% (Clegg and Brimblecombe, 1985). Apparent loss of Cl^- from aerosol samples has been frequently observed along the California coast (Jacob et al., 1985; Munger et al., 1988). Due to the reaction between $HNO_{3(g)}$ and $NaCl_{(s)}$, NO_3^- replaces Cl^- in the NaCl-dominated aerosol and releases $HCl_{(g)}$. The emitted gas-phase HCl should be included in the acid base balance because it is efficiently scavenged by cloud droplets. Infact, unlike the aerosol samples most of the cloudwater samples did not have a Cl^- deficit.

Size-fractionated Cloudwater Samples

Figure 14 illustrates the theoretical performance of the size-fractionating inlet. The initial droplet-size spectra are theoretical curves generated from Best's formula (1951). The fraction of the liquid water retained on the rods and the strands is calculated from impaction theory for droplets on a cylinder (Friedlander, 1977). Collection efficiency curves are shown for the strands and rods in Figure 14. The fraction of air sampled by the collector is a function the diameter and spacing of the collector elements as shown in the following relationship:

$$\mathbf{F}_{s} = 1 - [1 - \frac{\mathbf{D}_{c}}{\Delta \mathbf{C}}]^{n}$$

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where F_s is the fraction of incoming air that is sampled, D_c is the diameter of an individual collection element, ΔC is the spacing of the elements, and n is the number of rows. The CASC contains 6 rows of 510 μ m strands, spaced 1.8 mm apart, which yields an 86% sampling efficiency. The size-fractionating inlet contains 4 rows of 12.7 mm rods, spaced 25.4 mm apart, and samples 91% of the incoming air.

At low liquid water contents, where a higher proportion of the water is present in droplets too small to be collected by the rods, most of the water will be collected on the strands. With increasing LWC the mass median diameter increases and more sample is collected on the rods. Because the rods do not have a sharp lower size cut-off, there is considerable overlap in the portions of the droplet spectrum sampled by the rods and strands. Any differences observed in the composition of the two sample fractions, therefore, should be considered as a lower bound on the actual differences present if the droplets could be separated more cleanly.

Comparisons of the ionic concentrations in the two fractions collected during each interval suggest that there is a large difference between the average composition of the smaller droplets and that of the larger droplets. For every interval sampled, the concentration of Na⁺ and Mg²⁺ in the large droplet fraction was observed to be higher than in the small droplet fraction. With the exception of one very low concentration sample, the same was true for Ca^{2+} . Concentrations of SO_4^{2-} , NO_3^{-} , NH_4^+ , and H⁺ (Figure 9) were almost always higher in the small droplet fraction. Neither fraction had consistently higher Cl⁻ concentrations (Figure 10); Cl⁻ was nearly equal in both fractions. The Cl⁻:Na⁺ ratio in the two fractions differs, however. In the large–droplet fraction, Na⁺ and Cl⁻ are close to the seawater ratio, while the fine–droplet fraction has an excess of Cl⁻ relative to seawater. As noted above, Cl⁻ is removed from the sea salt aerosol by reaction with HNO₃. Gas–phase HCl should be scavenged equally by all droplets.

Comparison of the organic acid concentrations (Fig. 12) in the two fractions indicates that there was little difference; this result would be expected if the clouds were in equilibrium

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with the surrounding air and the droplet pH did not vary appreciably with size. The calculated equilibrium partial pressures of each fraction are comparable (i.e. $C_{aq} = K_{h,i}P_i$). Over the observed cloudwater pH range of 3 to 3.6, the majority of the organic acid will be in the gas phase (Munger et al., 1989). The behavior of HCOOH and CH₃COOH is discussed by Munger et al. (1989). In addition, the CH_2O concentrations in the initial fractionated samples are equivalent, which is also consistent with equilibrium considerations. However, beginning at 5:30 there was a spike in [CH₂O], [HCOOH], and [CH₃CO₂H] as well as the major inorganic ions. The organic acids appear to maintain their apparent equilbrium between the gas and aqueous phases, while the CH₂O showed a pronounced increase in the small-droplet fraction. This difference disappears over the subsequent three hours. The sharpness of the pulse suggests the passage of a plume or a major wind shift. The CH₂O data imply that the precursor nuclei for the smaller droplets contain formaldehyde that is not in equilibrium with the gas phase. The presence of α -hydroxymethanesulfonate (HMSA) and other aldehydebisulfite adducts could account for this apparent non-equilibrium behavior (Munger et al., 1986). At pH 3.5, the kinetics of aldehyde-bisulfite adduct dissociation is quite slow (Betterton et al., 1988), thus HMSA in the precursor aerosol would be retained in the droplet and measured analytically as CH_2O . The presence of detectable S(IV) during this period is consistent with this argument. Of special interest in this regard, S(IV) and H_2O_2 were not found to be present simultaneously in either size fraction.

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The differences in concentration between the size fractions are consistent with the suggestion that large nuclei produce large droplets and small nuclei produce small droplets (Best, 1951; Mason and Chien; 1962; Hudson, 1984; Jensen and Charlson, 1984; Noone et al., 1988). For the size-dependent aerosol composition to be preserved in the droplets requires nucleation scavenging to be the dominant scavenging process. Because Na⁺, Ca²⁺ and Mg²⁺ are associated with sea salt and soil dust (Seinfeld, 1986) they are found predominantly in the large droplets; while NH_4^+ , SO_4^{2-} , and to some extent NO_3^- , which are mostly found in secondary aerosol, would be in the small droplets (Seinfeld, 1986). However, NO_3^- , which can exist in the

gas phase or on large aerosol by exchange with Cl⁻, would also be found to a substantial extent in large droplets. However, the cloudwater concentrations of these ions may be altered by absorption of precursor gas phase species followed by chemical reaction. For example, $NH_{3(g)}$ can be absorbed by the droplets and protonated to form NH_4^+ ; $HNO_{3(g)}$ can be absorbed, followed by deprotonation to yield NO_3^- ; $SO_{2(g)}$ can be absorbed and oxidized to SO_4^{2-} . The first two processes are extremely rapid, while the oxidation of S(IV) to S(VI) in cloudwater is also rapid in the presence H_2O_2 or a metal catalyst (Hoffmann and Jacob, 1984). Since S(IV)and H_2O_2 were not found concurrently in the time- and size-resolved samples, the rapid oxidation of S(IV) to S(VI) by H_2O_2 was likely to have taken place (McArdle and Hoffmann, 1983).

Summary

Aerosol composition at elevated sites in the South Coast air basin was a mixture of sea salt and pollution-derived secondary aerosol. The influence of sea salt declined with increasing distance from the coast. Abundant HNO_3 reacted with the NaCl in sea-salt aerosol to give $HCl_{(g)}$ and NaNO₃ in the aerosol. At inland sites aerosol concentrations differ during onshore and offshore flow. The highest concentrations were observed during the day when the onshore flow transports pollutants to the sites, while lower concentrations were observed at night when drainage flows from nearby mountains influenced the sites. Variations in liquid water content are a major influence on cloudwater concentration. Steady declines in concentration were also observed during periods of steady LWC. These may be due to removal by drizzle, dilution by advection, or entrainment of cleaner air from aloft.

Differences in the composition of size-fractionated cloudwater samples suggest that large droplets are formed from sea salt and soil dust, which are large aerosol, and small droplets are formed on small secondary aerosol composed of ammonium sulfate and nitrate. Components that exist partly in the gas phase (e.g. Cl^- , HCOOH, and CH_3COOH) did not appear to be size segregated. The exchange of HNO₃ for HCl on sea salt provides a means for Cl^- to associate

with small droplets. Because Cl⁻ is removed from the sea salt aerosol during the day, the large droplets that form from them are deficient in Cl⁻. Chloride is retained in the air mass and some ends up in the small droplets that formed on secondary aerosol.

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San Pedro Hill															
	$\begin{array}{cccccccccccccccccccccccccccccccccccc$														
	11			_		Day	11		31			11			
$ \begin{array}{c} {\rm N} \\ {\rm Min} \\ {\rm Max} \\ {\rm Avg} \\ \sigma \end{array} $	$18 \\ 0 \\ 188 \\ 81 \\ 65$	$13 \\ 17 \\ 430 \\ 186 \\ 122$	$18 \\ 0 \\ 47 \\ 22 \\ 10$	18 0 49 19 16	$16 \\ 0 \\ 105 \\ 25 \\ 33$	18 7 147 69 37	$18 \\ 22 \\ 448 \\ 179 \\ 106$		18 0 105 19 28	$14 \\ 0 \\ 166 \\ 91 \\ 57$	$13 \\ 36 \\ 479 \\ 206 \\ 131$	14 7 279 164 74			
						Night									
$ \begin{array}{c} {\rm N} \\ {\rm Min} \\ {\rm Max} \\ {\rm Avg} \\ \sigma \end{array} $	$19 \\ 0 \\ 170 \\ 52 \\ 46$	$12 \\ 0 \\ 624 \\ 169 \\ 183$	19 0 56 22 12	19 0 44 15 13	16 0 90 34 31	$18\\6\\435\\127\\118$	$ 19 \\ 32 \\ 475 \\ 162 \\ 121 $		19 0 29 5 9	$17 \\ 0 \\ 124 \\ 28 \\ 31$	$10 \\ 71 \\ 635 \\ 212 \\ 180$	17 12 448 155 129			
								••••••		-					
	Na ⁺ NH ₄ ⁺ Ca ²⁺ Mg ²⁺ Cl ⁻ NO ₃ ⁻ SO ₄ ²⁻ NH ₃ HNO ₃ N(-3) N(V)														
	Na⁺ 	NH ⁺	Ca ²⁺	Mg²+ → n	Hen Cl ⁻ eq m ⁻³	ninger NO₃	SO ₄ ²⁻		NH₃ ───	HNO → nmol	3 N(-3) .e m ⁻³ ←	N(V)			
·	Na⁺ ├───	NH ⁺	Ca ²⁺	Mg ²⁺ ──→ n	Henn Cl ⁻ eq m ⁻³	NO₃ NO₃ ← Day	r Flats 504- 		NH ₃	HNO → nmol	3 N(-3) e m ⁻³ ←	N(V)			
N Min Max Avg σ	Na⁺ 9 0 134 59 46	NH ⁺ 9 4 838 367 284	Ca ²⁺ 9 0 93 39 31	$\begin{array}{c} Mg^{2+} \\ \longrightarrow n \\ 9 \\ 0 \\ 54 \\ 19 \\ 15 \end{array}$	Heni Cl ⁻ eq m ⁻³ 9 0 29 6 11	ninger NO₃	Flats SO ₄ ²⁻ 9 5 533 -255 195		NH ₃ 9 0 99 40 40	HNO \rightarrow nmol $\begin{pmatrix} 8\\11\\897\\363\\317 \end{pmatrix}$	3 N(-3) e m ⁻³ ← 9 4 916 407 310	N(V) 8 19 1255 526 450			
N Min Max Avg σ	Na⁺ 9 0 134 59 46	NH ₄ 9 4 838 367 284	Ca ²⁺ 9 0 93 39 31	$Mg^{2+} \longrightarrow n$ g 0 54 19 15	Heni Cl ⁻ eq m ⁻³ 9 0 29 6 11	NO ₃ NO ₃ Day 9 7 442 173 136 Night	Flats SO ₄ ²⁻ 9 5 533 255 195		NH ₃ 9 0 99 40 40	HNO → nmol 8 11 897 363 317	${}_{3} N(-3)$ e m ⁻³ \leftarrow 9 4 916 407 310	N(V) 8 19 1255 526 450			

Table 1. Summary Statistics for Aerosol Samples Collected at Three Elevated in the Los Angeles Basin (6–13 to 7–17–88)

- 19-

Table 1. (continued)

Kellogg Hill

	Na⁺	NH_4^+	Ca ²⁺	Mg ²⁺	Cl-	NO_3^-	SO_4^{2-}	NH ₃	HNO	3 N(-3)	N(V)
	I 			ne	eq m⁻₃	~	11		→ nmoi	e m⁻₃ ←]]
						Day					
N Min Max Avg σ	$7\\8\\149\\92\\48$	7 9 875 575 302	$7\\62\\270\\162\\60$	$7 \\ 12 \\ 62 \\ 44 \\ 16$	$7 \\ 0 \\ 16 \\ 5 \\ 6$	$7\\10\\629\\413\\204$	$7 \\ 7 \\ 451 \\ 296 \\ 156$	7 0 339 140 120	$7 \\ 6 \\ 624 \\ 277 \\ 185$	$7\\8\\1063\\714\\353$	$7\\16\\1191\\690\\358$
	·					Night					
N Min Max Avg σ	$11 \\ 44 \\ 183 \\ 112 \\ 43$	$11 \\ 139 \\ 594 \\ 343 \\ 167$	$11 \\ 40 \\ 109 \\ 75 \\ 21$	$11 \\ 11 \\ 61 \\ 33 \\ 16$	$11 \\ 0 \\ 72 \\ 20 \\ 24$	$11 \\ 91 \\ 452 \\ 266 \\ 118$	11 94 372 201 80	11 0 283 89 82	$ \begin{array}{r} 11 \\ 9 \\ 63 \\ 40 \\ 19 \\ \end{array} $	$11 \\ 279 \\ 620 \\ 432 \\ 128$	11 133 512 306 128

	V(mL)	pН	Na⁺	NH_4^+	Ca^{2+}	Mg ²⁺	Cl-	NO_3^-	SO_4^{2-}	$\rm CH_2O$	-/+	LWC
						µN ←				$\mu \mathbf{M}$		g m ⁻³
	•				14	June 19	87					
N Min Max Avg Vol Wt.	15 18 68 48 Avg	$15 \\ 2.80 \\ 3.28 \\ 3.02 \\ 3.02 \\ 3.02$	15 149 734 506 468	$15 \\ 323 \\ 744 \\ 501 \\ 466$	15 15 88 58 54	15 47 214 149 139	15 247 787 538 502	1570216081062991	$15 \\ 574 \\ 1407 \\ 939 \\ 880$	0	15 1.09 1.19 1.14	$15 \\ 0.02 \\ 0.11 \\ 0.07 \\ 0.06$
					19 .	June 19	87					
N Min Max Avg Vol Wt.	3 26 36 33 Avg	$3 \\ 3.18 \\ 3.33 \\ 3.25 \\ 3.25 \\ 3.25$	3 3258 3796 3486 3553	3 665 1237 909 980	$3 \\ 281 \\ 460 \\ 353 \\ 375$	$3 \\ 593 \\ 731 \\ 652 \\ 670$	$\begin{array}{c} 3 \\ 1654 \\ 1701 \\ 1676 \\ 1682 \end{array}$	$\begin{array}{c} 3 \\ 1281 \\ 2137 \\ 1674 \\ 1784 \end{array}$	$\begin{array}{c} 3\\978\\1131\\1078\\1102\end{array}$	$3 \\ 25 \\ 31 \\ 27 \\ -$	$3 \\ 0.73 \\ 0.75 \\ 0.74 $	$3 \\ 0.02 \\ 0.06 \\ 0.04 \\ 0.04$
					20	June 19	87					
N Min Max Avg Vol Wt.	6 9 65 48 Avg	6 3.11 3.35 3.24 3.24	5 1722 2574 2024 1812	6 347 605 478 470	4 173 272 225 127	6 388 715 476 444	$\begin{array}{r} 6 \\ 1379 \\ 1756 \\ 1468 \\ 1434 \end{array}$	$\begin{array}{c} 6 \\ 498 \\ 1402 \\ 850 \\ 815 \end{array}$	6 749 1290 1032 964	4 21 31 24	6 0.73 1.84 1.06	$\begin{array}{c} 6 \\ 0.03 \\ 0.10 \\ 0.06 \\ 0.06 \end{array}$
					21	June 19	87					
N Min Max Avg Vol Wt.	3 12 66 47 Avg	3 3.48 3.55 3.52 3.51	$\begin{array}{c} 3\\706\\995\\841\\866\end{array}$	3 169 295 228 238	$3 \\ 79 \\ 142 \\ 108 \\ 113$	$ \begin{array}{r} 3 \\ 192 \\ 253 \\ 215 \\ 216 \end{array} $	3 735 837 793 787	3 377 915 590 606	3 312 447 373 382	$\begin{array}{c}2\\12\\14\\13\\\end{array}$	3 1.00 1.07 1.04	$\begin{array}{c} 3 \\ 0.03 \\ 0.17 \\ 0.09 \\ 0.09 \end{array}$
					22	June 19	87					
N Min Max Avg Vol Wt.	9 13 62 56 Avg	9 2.84 3.43 3.21 3.23	$9 \\ 206 \\ 1257 \\ 462 \\ 424$	$9 \\ 311 \\ 1126 \\ 552 \\ 467$	9 27 243 79 70	9 53 311 120 110	9 267 978 485 447	9 436 1632 827 704	$9 \\ 354 \\ 1478 \\ 641 \\ 540$	7 11 27 16	9 0.94 1.09 1.01	$9 \\ 0.04 \\ 0.12 \\ 0.08 \\ 0.07$

Table 2. Concentrations in Cloudwater Samples Collected atSan Pedro Hill (6–13 to 7–17–87).

Table 2. (continued)

	V(mL) pH	Na ⁺	NH_4^+	Ca^{2+}	Mg^{2+}	Cl-	NO_3^-	SO_4^{2-}	CH_2C) -/+	LWC
						• µN ←				$\mu \mathbf{M}$		g m ⁻³
					23	June 1	987					
N Min Max Avg Vol Wt.	20 61 66 62 Avg	$20 \\ 3.15 \\ 3.62 \\ 3.47 \\ 3.49$	$20\\123\\452\\200\\185$	20 223 528 293 277	20 17 95 38 34	20 33 131 55 50	20 173 505 257 241	20 277 1024 407 371	$20 \\ 224 \\ 608 \\ 342 \\ 324$	0	20 0.95 1.27 1.08	$\begin{array}{c} 20 \\ 0.02 \\ 0.16 \\ 0.12 \\ 0.10 \end{array}$
					24	June 1	987					
N Min Max Avg Vol Wt.	24 17 66 56 . Avg	$\begin{array}{c} 24 \\ 2.53 \\ 2.94 \\ 2.76 \\ 2.78 \end{array}$	24 189 3148 794 553	24 462 2859 1495 1 340	20 30 560 134 99	$24 \\ 56 \\ 601 \\ 179 \\ 133$	$\begin{array}{c} 24 \\ 309 \\ 1613 \\ 666 \\ 552 \end{array}$	$\begin{array}{c} 24 \\ 1291 \\ 5623 \\ 2627 \\ 2226 \end{array}$	$\begin{array}{c} 24 \\ 1119 \\ 3246 \\ 1871 \\ 1669 \end{array}$	22 18 51 34	24 1.09 1.51 1.18	$\begin{array}{c} 22 \\ 0.02 \\ 0.25 \\ 0.11 \\ 0.09 \end{array}$
					25	June 1	987					
N Min Max Avg Vol Wt.	23 10 68 42 . Avg	$\begin{array}{c} 23 \\ 2.63 \\ 3.11 \\ 2.78 \\ 2.81 \end{array}$	$23 \\ 142 \\ 1618 \\ 468 \\ 323$	23 410 9383 1702 1072	$23 \\ 30 \\ 1143 \\ 138 \\ 73$	$23 \\ 37 \\ 1151 \\ 135 \\ 73$	$23 \\ 132 \\ 2096 \\ 402 \\ 254$	23 832 8191 2708 2062	$23 \\ 660 \\ 5796 \\ 1874 \\ 1365$	$ \begin{array}{c} 15\\ 16\\ 37\\ 26\\ \hline \end{array} $	23 0.89 1.42 1.18	$\begin{array}{c} 21 \\ 0.00 \\ 0.34 \\ 0.18 \\ 0.11 \end{array}$
					26	June 1	987					
N Min Max Avg Vol Wt	21 32 69 55 . Avg	$\begin{array}{c} 21 \\ 2.99 \\ 3.51 \\ 3.31 \\ 3.34 \end{array}$	$21 \\ 28 \\ 315 \\ 89 \\ 67$	$21 \\ 423 \\ 1979 \\ 1058 \\ 931$	$21 \\ 8 \\ 164 \\ 27 \\ 20$	$21 \\ 10 \\ 86 \\ 25 \\ 19$	$21 \\ 43 \\ 223 \\ 90 \\ 71$	$21 \\ 454 \\ 2462 \\ 1044 \\ 915$	$21 \\ 387 \\ 1971 \\ 794 \\ 660$	0	$21 \\ 0.79 \\ 1.36 \\ 1.11 \\$	$19 \\ 0.08 \\ 0.29 \\ 0.21 \\ 0.21$
					0 1	t July 1	987					
N Min Max Avg Vol Wt	6 8 29 15 Avg	$\begin{array}{c} 6\\ 3.15\\ 3.23\\ 3.18\\ 3.18\\ 3.18\end{array}$	$7 \\ 336 \\ 912 \\ 480 \\ 440$	$7 \\ 242 \\ 638 \\ 517 \\ 555$	$7 \\ 57 \\ 215 \\ 111 \\ 108$	$7 \\ 90 \\ 248 \\ 129 \\ 120$	$7\\339\\745454415$	$7\\401\\1138\\741\\754$	$7\\435\\836\\652\\671$	0		$\begin{array}{c} 6 \\ 0.02 \\ 0.15 \\ 0.10 \\ 0.04 \end{array}$

Table 2. (continued)

	V(mL) pH	Na ⁺	NH_4^+	Ca^{2+}	Mg^{2+}	Cl-	NO_{3}^{-}	SO_4^{2-}	CH_2O	-/+	LWC
					<u> </u>	• μN ←				$\mu \mathbf{M}$		g m ⁻³
					03	3 July 1	987					
N Min Max Avg Vol Wt.	14 20 62 55 Avg	$14 \\ 2.42 \\ 3.30 \\ 2.72 \\ 2.66$	$14 \\ 308 \\ 683 \\ 443 \\ 415$	$14\\394\\2770\\1652\\1675$	14 71 286 128 119	$14\\83\\202\\124\\116$	$14 \\ 327 \\ 656 \\ 466 \\ 445$	$14 \\ 535 \\ 4656 \\ 2987 \\ 2994$	14 492 3028 1900 1892	0	$14 \\ 0.98 \\ 1.26 \\ 1.16 $	$14 \\ 0.02 \\ 0.11 \\ 0.07 \\ 0.06$
					0	7 July 1	987					
N Min Max Avg Vol Wt.	7 23 66 55 Avg	$7 \\ 3.14 \\ 3.51 \\ 3.33 \\ 3.31 \\$	$7 \\ 235 \\ 719 \\ 489 \\ 412$	$7 \\ 266 \\ 1108 \\ 473 \\ 470$	$7 \\ 28 \\ 127 \\ 57 \\ 52$	7 53 156 107 91	7 284 821 537 453	7 332 1301 706 713	7 384 810 562 555	$ \begin{array}{c} 7\\12\\22\\16\\\end{array} $	7 1.07 1.19 1.13	$7 \\ 0.03 \\ 0.21 \\ 0.12 \\ 0.10$
					0	8 July 1.	987					
N Min Max Avg Vol Wt.	9 4 65 53 Avg	9 2.81 3.91 3.09 2.99	$9\\59\\1365\\485\\481$	$9\\88\\2458\\866\\855$	$9 \\ 20 \\ 508 \\ 130 \\ 119$	9 15 335 117 116	$9\\71\\1055\\415\\409$	$9 \\ 172 \\ 3419 \\ 1435 \\ 1435 \\ 1435 \end{cases}$	$9\\143\\2647\\1044\\1052$	0 	9 1.07 1.27 1.15	$9 \\ 0.00 \\ 0.25 \\ 0.13 \\ 0.11$
					1.	3 July 1	987					
N Min Max	2 9 36	$2 \\ 3.79 \\ 3.83$	$2 \\ 204 \\ 223$	$\begin{array}{c}2\\210\\240\end{array}$	$2 \\ 45 \\ 52$	$2 \\ 57 \\ 57 \\ 57$	$\begin{array}{c}2\\242\\264\end{array}$	$2 \\ 174 \\ 217$	2 230 230	0 10	$2 \\ 0.93 \\ 1.10$	$\begin{array}{c}2\\0.01\\0.05\end{array}$
					1.	4 July 1	987					
N Min [.] Max Avg Vol Wt.	21 29 65 40 Avg	$21 \\ 3.13 \\ 3.91 \\ 3.61 \\ 3.63$	$21 \\ 64 \\ 207 \\ 106 \\ 97$	$\begin{array}{c} 21 \\ 129 \\ 977 \\ 351 \\ 295 \end{array}$	$21 \\ 13 \\ 55 \\ 26 \\ 22$	21 19 59 29 26	$21 \\ 95 \\ 240 \\ 146 \\ 138$	$\begin{array}{c} 21 \\ 112 \\ 600 \\ 285 \\ 235 \end{array}$	$21 \\ 182 \\ 820 \\ 382 \\ 319$	$ \begin{array}{c} 11 \\ 5 \\ 38 \\ 15 \\ \end{array} $	21 0.94 1.17 1.02	$19 \\ 0.12 \\ 0.31 \\ 0.22 \\ 0.21$
					1.	4 July 1	987					
N Min Max Avg Vol Wt	19 37 42 39 . Avg	$\begin{array}{c} 21 \\ 3.23 \\ 3.73 \\ 3.50 \\ 3.52 \end{array}$	$21 \\ 40 \\ 267 \\ 160 \\ 175$	$21 \\ 131 \\ 391 \\ 227 \\ 210$	$21 \\ 10 \\ 77 \\ 30 \\ 29$	$21 \\ 13 \\ 65 \\ 38 \\ 40$	$21 \\ 57 \\ 272 \\ 162 \\ 173$	$21 \\ 167 \\ 602 \\ 307 \\ 277$	$\begin{array}{c} 21 \\ 212 \\ 550 \\ 327 \\ 311 \end{array}$	8 6 15 10	21 0.95 1.08 1.01	$19 \\ 0.05 \\ 0.23 \\ 0.17 \\ 0.14$

Table 2. (continued)

	V(mL) pH	Na^+	NH_4^+	Ca^{2+}	Mg^{2+}	Cl-	NO_3^-	$\mathrm{SO}_4^{2\text{-}}$	CH_2O) -/+	LWC
				<u> </u>		• μN ←				$\mu \mathbf{M}$		g m ⁻³
					16	July 1	987					
N Min Max Avg Vol Wt	$16 \\ 12 \\ 68 \\ 51 \\ . Avg$	$16 \\ 2.64 \\ 3.93 \\ 3.12 \\ 3.06$	$16 \\ 22 \\ 271 \\ 119 \\ 93$	16 328 2161 727 639	$16 \\ 14 \\ 236 \\ 110 \\ 87$	$16 \\ 8 \\ 100 \\ 43 \\ 35$	16 40 357 155 131	16 190 2797 854 819	16 219 2438 992 888	0	$16 \\ 0.93 \\ 1.12 \\ 1.06$	$15 \\ 0.02 \\ 0.24 \\ 0.14 \\ 0.11$
					17	7 July 1:	987					
N Min Max Avg Vol Wt	20 25 56 49 . Avg	$\begin{array}{c} 20 \\ 3.50 \\ 4.98 \\ 4.09 \\ 3.94 \end{array}$	20 6 92 35 28	$20 \\ 44 \\ 504 \\ 136 \\ 101$	$9 \\ 3 \\ 47 \\ 14 \\ 6$	$9 \\ 1 \\ 28 \\ 10 \\ 4$	$20 \\ 11 \\ 98 \\ 43 \\ 35$	$20 \\ 28 \\ 300 \\ 125 \\ 100$	20 39 376 161 138	0	20 0.92 1.23 1.09	$18 \\ 0.05 \\ 0.26 \\ 0.16 \\ 0.14$
				Al	l Sample	es 6–14	to 7—17	<i>–88</i>				
N Min Max Avg Vol Wt	240 1 69 49 . Avg	$242 \\ 2.42 \\ 4.98 \\ 3.25 \\ 3.15$	$241 \\ 6 \\ 3796 \\ 404 \\ 263$	242 44 9383 775 632	$225 \ 3 \ 1143 \ 82 \ 51$	$231 \\ 1 \\ 1151 \\ 107 \\ 65$	$242 \\ 11 \\ 2096 \\ 369 \\ 256$	$242 \\ 28 \\ 8191 \\ 1185 \\ 941$	$242 \\ 39 \\ 5796 \\ 917 \\ 736$	80 5 51 23	.241 0.73 1.84 1.09	$\begin{array}{c} 227 \\ 0.00 \\ 0.34 \\ 0.14 \\ 0.103 \end{array}$

Table 3. Concentrations of Organic Acids, S(IV), HCHO, and H_2O_2

Cloudwater Samples Collected at San Pedro Hill

V(mL) pH	Time (hrs)	S(IV) CH ₂ O H ₂ O ₂ HCO ₂ H* CH ₃ COOH*	+/-
		$ \longrightarrow \mu M \longleftarrow $	

7-15-88 Size-fractionated Sample from the Front Rods of the CASC

68	3 34	4.33-5.00	0	10	10	10	11	1.08
71	3.27	5:00-5:30	ŏ	16	Ô	$\frac{10}{21}$	13	1.00
71	3.26	5:30-6:00	Ō	27	Ō	40	19	1.00
74	3.29	6:00-6:30	6	29	0	37	18	0.98
112	3.42	6:30-7:00	6	16	0	26	11	1.02
121	3.42	7:00-7:30	6	13	0	17	9	1.04
136	3.44	7:30-8:00	0	13	4	19	9	0.99
122	3.49	8:00-8:30	0	13	10	18	10	1.02
116	3.52	8:30-9:00	0	13	16	20	9	1.02
64	3.40	10:30-11:00	0	18	49	33	13	1.06

7–15–88 Size-fractionated Sample from the Back Strings of the CASC

29	3.42	4:33-5:00	0	11	9	16	10	1.03
28	3.28	5:00-5:30	0	13	0	18	11	1.05
31	3.10	5:30-6:00	6	26	0	34	17 .	0.99
28 [°]	3.08	6:00-6:30	10	55	0	38	22	0.98
38	3.24	6:30 - 7:00	5	25	0	32	20	0.98
39	3.35	7:00-7:30	0	17	0	18	9	1.02
38	3.38	7:30-8:00	0	16	3	19	9	0.96
30	3.42	8:00-8:30	0	17	6	19	10	1.01
32	3.47	8:30-9:00	0	14	11	23	10	1.02
30	3.25	10:30-11:00	0	21	39	40	17	1.06

Samples Collected from 7-7-87 to 7-14-87

Parameter	V(m	L) pH	S(IV)	CH_2	O H ₂ O ₂	HCO	OH∗CH ₃ COO	H
					$\longrightarrow \mu \mathbf{M}$	4	[]	
Ν	21	21		20	16	21	21	
Min	9	3.14		5	4	12	6	
Max	59	3.83		38	72	43	31	
Avg.	38	3.58		13	42	20	10	

* This represents the total RCOOH (i.e. $[RCOOH] + [RCO_2])$ unless otherwise noted.

	V(mL)) pH	Na ⁺	NH_4^+	Ca ²⁺	Mg^{2+}	Cl-	NO_3^-	SO_4^{2-}	CH_2O	-/+	LWC
						→ μN ←				$\mu \mathbf{M}$		g m ⁻³
					20	June 1	987					
N Min Max Avg Vol Wt.	6 24 65 48 Avg	$\begin{array}{c} 6 \\ 2.75 \\ 3.59 \\ 3.15 \\ 3.09 \end{array}$	$\begin{array}{c} 6 \\ 180 \\ 1652 \\ 542 \\ 442 \end{array}$	$\begin{array}{c} 6\\ 805\\ 3509\\ 1575\\ 1418 \end{array}$	$\begin{array}{c} 6\\ 39\\ 454\\ 136\\ 106 \end{array}$	$6\\48\\435\\144\\118$	6 169 975 377 327	$\begin{array}{r} 6\\ 877\\ 4034\\ 1879\\ 1715 \end{array}$	$\begin{array}{r} 6 \\ 447 \\ 2028 \\ 885 \\ 796 \end{array}$	6 66 91 76 78	6 0.90 1.04 0.99	$\begin{array}{c} 6 \\ 0.03 \\ 0.09 \\ 0.05 \\ 0.05 \end{array}$
					21	June 1	987					
N Min Max Avg Vol Wt.	12 7 45 39 Avg	$12 \\ 2.62 \\ 3.75 \\ 3.01 \\ 2.96$	$12 \\ 183 \\ 1553 \\ 485 \\ 340$	$\begin{array}{c} 12 \\ 603 \\ 3172 \\ 1583 \\ 1368 \end{array}$	12 42 520 138 89	12 49 400 129 90	$12 \\ 161 \\ 666 \\ 308 \\ 259$	$12 \\ 787 \\ 5472 \\ 2116 \\ 1773$	$12 \\ 36 \\ 2606 \\ 1084 \\ 963$	$11 \\ 53 \\ 109 \\ 68 \\ 62$	12 0.76 1.09 0.98	$12 \\ 0.01 \\ 0.18 \\ 0.08 \\ 0.05$
					10	July 19	987					
N Min Max avg Vol Wt.	20 29 63 37 Avg	20 2.88 3.88 3.24 3.22	20 48 195 82 68	$\begin{array}{c} 20 \\ 671 \\ 1760 \\ 1022 \\ 864 \end{array}$	20 47 167 91 76	20 20 99 36 29	$20 \\ 91 \\ 214 \\ 128 \\ 111$	20 735 2065 1076 949	$20 \\ 368 \\ 1094 \\ 589 \\ 526$	0	20 0.81 1.08 0.96	19 0.01 0.17 0.10 0.05
					10	6 July 1	987					
N Min Max Avg Vol Wt	20 9 65 58 Avg	$\begin{array}{c} 20 \\ 2.68 \\ 4.78 \\ 3.36 \\ 3.48 \end{array}$	$20 \\ 5 \\ 149 \\ 43 \\ 27$	$\begin{array}{c} 20 \\ 324 \\ 4085 \\ 1392 \\ 740 \end{array}$	20 7 397 80 27	20 2 77 23 10	$20 \\ 18 \\ 115 \\ 48 \\ 35$	$20 \\ 284 \\ 1908 \\ 804 \\ 525$	$\begin{array}{c} 20 \\ 137 \\ 1474 \\ 549 \\ 346 \end{array}$	0	$20 \\ 0.46 \\ 1.33 \\ 0.78 $	$18 \\ 0.01 \\ 0.40 \\ 0.24 \\ 0.14$
					17	7 June 1	987					
N Min Max Avg Vol Wt.	18 40 65 62 Avg	$18 \\ 3.03 \\ 3.94 \\ 3.35 \\ 3.31$	$18 \\ 3 \\ 273 \\ 55 \\ 46$	$18 \\ 273 \\ 2435 \\ 817 \\ 752$	$18 \\ 15 \\ 985 \\ 171 \\ 136$	$18 \\ 7 \\ 171 \\ 40 \\ 34$	$18 \\ 22 \\ 301 \\ 73 \\ 63$	18 231 3319 967 897	$18 \\ 159 \\ 2028 \\ 628 \\ 572$	0	18 0.95 1.18 1.05	$17 \\ 0.07 \\ 0.23 \\ 0.17 \\ 0.15$

Table 4. Concentrations in Cloudwater Samples Collected at Henninger Flats during the Period of 6–20 to 7–17–87.

Table 4. (continued)

	V(mI	L) pH	Na ⁺	$\rm NH_4^+$	Ca^{2+}	Mg^{2+}	Cl-	NO_3^-	SO_4^{2-}	CH_2C	-/+	LWC
						• µN ←				$\mu \mathbf{M}$		g m ⁻³
				All	Sampl	es 6–20	to 7—1'	788				
N	76	76	76	76	76	76	76	76	76	17	76	72
Min	7	2.62	3	273	7	2	18	231	36	53	0.46	0.01
Max	65	4.78	1652	4085	985	435	975	5472	2606	109	1.33	0.40
Avg	50	3.25	166	1203	118	57	142	1206	689	71	0.94	0.14
VoľWt	Avo	3 29	82	845	74	31	86	855	511			0.09



Figure 1

Map of the South Coast Air Basin. Major freeways and selected cities are shown. Sampling sites are indicated by •.



Figure 2

Diagram of the automated cloudwater collector system. Depicted in the figure are the CASC, a cloudwater sensor, and the fractionating collector, housed in a refrigerator.



Figure 3

A) Cl-concentrations in aerosol samples, plotted against Na⁺. --indicates the sea-salt ratio. B) Same as A, but for cloudwater.



Figure 4

A) The sum, $NO_3^- + SO_4^{2-}$, plotted vs NH_4^+ in aerosol samples. B) Total anions (Cl⁻ + $NO_3^- + SO_4^{2-} + HNO_3$) plotted vs total cations (Na⁺ + Ca²⁺ + Mg²⁺ + NH₄⁺ + NH₃) in aerosol samples



Figure 5

Concentrations of major ions and sea salts and LWC in cloudwater samples collected at San Pedro Hill on June 23 and 24, 1987. LWC is estimated from the sample collection rate and its theoretical efficiency.


Figure 6

Concentrations of major ions and sea salts and LWC in cloudwater samples collected at San Pedro Hill over the period July 14 – July 16, 1987. LWC is estimated from the sample collection rate and its theoretical efficiency.



Concentrations of major ions and sea salts and LWC in cloudwater samples collected at Henninger Flats on July 10, 1987. LWC is estimated from the sample collection rate and its theoretical efficiency.



Concentrations of major ions and sea salts and LWC in cloudwater samples collected at Henninger Flats on July 16, 1987. LWC is estimated from the sample collection rate and its theoretical efficiency.



Figure 9

Concentrations of major ions in size-fractionated cloudwater samples collected at San Pedro Hill on the morning of July 15, 1987.



Figure 10 Concentrations of sea salts in size–fractionated cloudwater samples collected at San Pedro Hill on the morning of July 15, 1987.



Collection rate for size-fractionated cloudwater samples collected at San Pedro Hill on the morning of July 15, 1987.



Figure 12

Formate (A), acetate (B), and formaldehyde (C) in size-fractionated cloudwater samples from San Pedro Hill. Large droplets are collected in the front fraction; are collected in the back fraction.



Figure 13

Theoretical collection efficiency vs droplet diameter for collection on the rods of the fractionating inlet and the strands of the CASC.







Collection of water on the fractionating inlet rods and CASC strands under three LWC conditions. Initial droplet-size distributions are given by Best's parameterized curves.