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ELSEVIER

Atmospheric Research 62 (2002) 1–10

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Measurements of selected dicarboxylic acids in marine cloud water

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Received 27 September 2001; received in revised form 4 February 2002; accepted 6 February 2002

Abstract

Measurements of selected dicarboxylic acid concentrations in cloud water from marine stratocumulus off of the central California coast are reported. The concentrations are consistent with previously reported concentrations in marine aerosols and suggest at least a partial anthropogenic origin. The concentrations were not sufficient to impact cloud drop activation and thus provide no support for an effect on cloud albedo, though such an effect cannot be ruled out in other circumstances. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Aerosols; Dicarboxylic acids; Activation

1. Introduction

The role of organic compounds in aerosol chemistry has been investigated, in a somewhat desultory fashion, for many years (cf., Cautreels and Van Cauwenberghe, 1976; Grosjean et al., 1978; Graedel and Goldberg, 1983; Simoneit, 1986; Rivera-Carpio et al., 1996; etc.), the end result of which has been a realization that organics constitute a very significant fraction of the atmospheric aerosol mass and play a large role in many atmospheric processes. More recently, attention has focused on soluble organics which, due to the difficulty of analysis, had not been much examined in earlier studies (Saxena and Hildemann, 1996). Of these, an important component is the carboxylic acids which have been found at significant levels in both pollution-derived and remote aerosols (e.g.,

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Meszaros et al., 1997; Limbeck and Puxbaum, 1999; Kawamura and Sakaguchi, 1999), and in clouds and precipitation (e.g., Kawamura and Kaplan, 1983; Weathers et al., 1988; Munger, 1989; Limbeck and Puxbaum, 2000). Recent studies have suggested that dicarboxylic acids in particular may be an important compound class, active as a component of cloud condensation nucleus (CCN) mass (Sempere and Kawamura, 1994; Corrigan and Novakov, 1999; Cruz and Pandis, 1997). Indeed, several studies have suggested that dicarboxylic acids, many of which are only sparingly soluble in water, could have an important influence on the CCN activation process (e.g., Shulman et al., 1996; Chuang et al., 1997; Laaksonen et al., 1998) and thus, potentially, on cloud albedo.

As of yet, however, only limited data on dicarboxylic acid concentrations in the marine atmosphere are available. Kawamura and Sakaguchi (1999) have reported measurements of aerosol dicarboxylic acid concentrations in the marine surface layer (from ship) over the Pacific Ocean which suggest that as much as 15% of the aerosol organic carbon mass is in the form of these diacids (~ 1% of the total aerosol mass), with oxalic predominating. These authors attribute the diacids in part to secondary production from lipids emitted from the ocean surface in salt particles and in part to long-range transport of pollution. No measurements have yet been reported at higher altitude and none to our knowledge in marine clouds, though some measurements in precipitation have been obtained (Sempere and Kawamura, 1996).

To at least partially rectify this lack of data, we report here measurements of both low molecular weight mono and dicarboxylic acids in stratiform clouds off the central California coast (flights centered on Monterey) obtained during the summer of 1999 (see Fig. 1). While convenient, this venue has the added advantage of representing a coastal marine area commonly impacted by anthropogenic pollution. Such areas have been touted as favorable for the significant influence of sparingly soluble organics on cloud processes (e.g., Laaksonen et al., 1998). Furthermore, this area is well known for the high susceptibility of its clouds to aerosol impact (Platnick and Twomey, 1994).

2. Methodology

All of the data presented here were obtained from the Twin Otter research aircraft operated by the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) based in Monterey, CA (Bluth et al., 1996). This aircraft was equipped with standard meteorological and cloud physics instruments including PMS particle spectrometers, temperature and dew point sensors, a gust probe, a Gerber Research cloud water sensor, MOUDI impactors and/or filter packs and a number of research aerosol sizing instruments built and maintained by the California Institute of Technology. Of particular note for this study, cloud water samples were obtained with a modified version of the well-known Mohnen slotted-rod cloud water collector (Mohnen, 1980; Hegg and Hobbs, 1986). Cloud water samples were tested for pH immediately after collection, treated with a biocide (chloroform) to prevent bacterial destruction of organics, and then stored at a nominal 5 °C until analysis by ion chromatography. Just prior to analysis, the samples were again tested for pH. A handful of aerosol filter samples (5) were also obtained for comparison with the cloud water samples. The filters utilized were 47 µm Teflon filters which were later

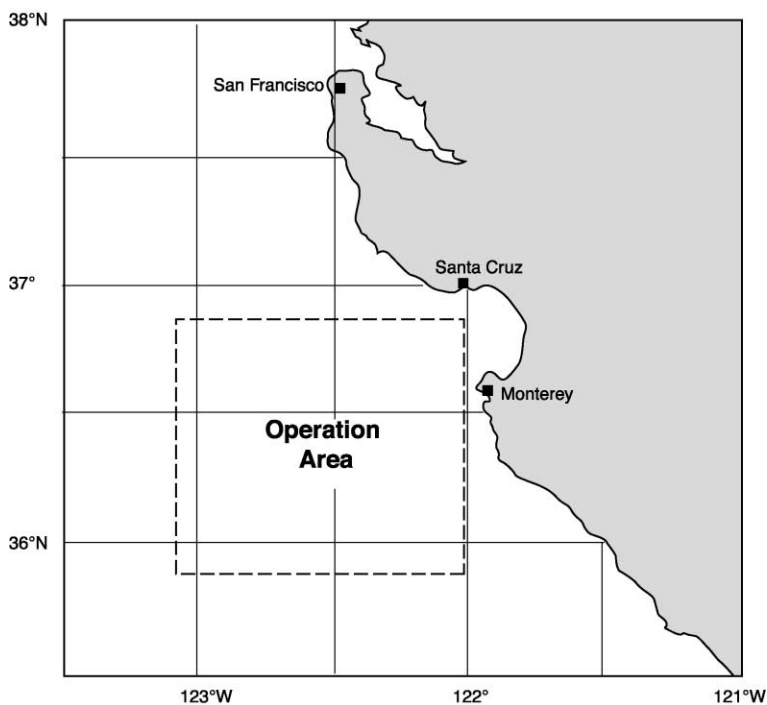


Fig. 1. Map of California coast showing the experiment operations area.

extracted in 10 ml DDI water (18 M Ω) and analyzed in the same IC as the cloud water samples (Fig. 2).

The ion chromatographic system utilized to analyze both the cloud water and filter extracts was a DIONEX Model 500 IC with an AS11 column and associated suppression and guard columns. Gradient elution was employed, with an eluent ramping from 0.5 to 8 mM NaOH over a period of 30 min. Filtered aerosol detection limits were derived from field blanks, while cloud water detection limits were derived from both standards and the noise determined from analyzing rinses of the cloud water collector with 18 M Ω water.

3. Data

The clouds sampled were all marine stratocumulus characteristic of the California coast during the summer months. Such clouds were the subject of the Monterey Area Ship Tracks experiment (MAST), which was in fact carried out in precisely the area of this experiment. The reader is referred to the MAST literature for more information on these clouds (e.g., Durkee et al., 2000). The cloud water samples (typically 5 ml) were all obtained near the cloud liquid water maximum, commonly within 100 m of cloud top. Roughly 5 min were necessary to collect each sample. On the basis of the wind direction, the samples for the 21st

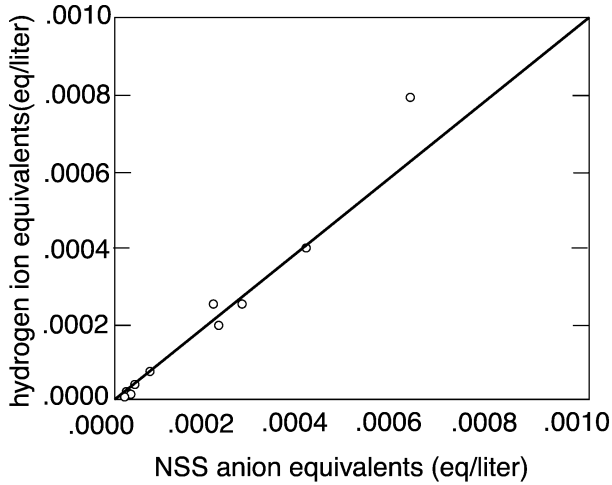


Fig. 2. Plot of the measured H^+ concentration against the sum of non-sea-salt anion equivalents (see text) for the 12 cloud water samples obtained.

and 24th should be essentially background marine while the two latter flights had an anthropogenic component as well.

Analyses of the cloud water data are given in Table 1. The first test of consistency for the data would normally be an ion balance. However, the only cation for which data is available is H^+ . Hence, we perform a modified ion balance, essentially a non-sea salt ion balance, by assuming that all of the Cl^- is derived from sea salt and this is neutralized by Na^+ . We further derive non-sea-salt (NSS) sulfate from the measured Cl^- concentration and the known ratio of chlorine to sulfate in seawater—essentially assuming no chlorine loss. Finally, given the pHs for the second dissociation for the organic diacids and the measured pHs, we assume that only oxalate is subject to a second proton dissociation in solution (its degree of dissociation being determined by the pH in the normal manner). With these assumptions, the ion balance was calculated and is depicted graphically in Fig. 1. Linear regression of H^+ onto the sum of NSS anion equivalents yield an R^2 of 0.97. With the regression line forced through zero, the slope is 1.15 ± 0.05 , suggesting a modest average 15% deficit of anions, possibly due to an unanalyzed organic acid anion such as malate or citrate, which are known to be present in sea water (cf., Creac'h, 1955).

The relative contribution of the various acids to the total acidity are interesting. Based on the approximate NSS ion balance just discussed, the dicarboxylic acids contribute an average 4% of the total acidity, while the two monocarboxylic acids contribute about 8% on average. However, this average is strongly influenced by a single sample (sample 1 on 21 June 1999) for which the monoacids contribute 44% of the total acidity. This sample has the lowest total NSS ion concentration, by far the highest pH and essentially no NSS sulfate. In other words, it is a true marine background sample, and an example of the high contribution to total acidity that acetic and formate acid are known to make under background conditions (cf., Keene et al., 1983). Exclusive of this sample, the monocarboxylic acids contribute

Table 1
Concentration of carboxylic acids found in the cloud water samples

Date (1999)	Sample no.	Cl ⁻	NO ₃ ⁻	SO ₄ ⁻	Acetate	Formate	Oxalate	Glutarate	Succinate	Malonate	LWC	PH	UV
June 21	1.000	202.800	6.000	23.000	2.080	2.010	0.480	0.138	0.000	0.290	0.160	5.400	24.000
June 21	2.000	83.900	5.000	23.000	0.000	0.540	0.510	0.019	0.000	0.290	0.150	4.700	24.000
June 21	3.000	101.500	6.000	23.000	0.140	0.600	0.580	0.017	0.000	0.350	0.170	4.600	21.000
June 24	1.000	55.700	4.000	15.000	0.960	0.890	0.340	0.060	0.000	0.720	0.220	4.600	13.000
June 24	2.000	35.200	14.000	26.000	0.990	1.010	0.670	0.168	0.280	1.300	0.170	4.100	16.000
June 24	3.000	27.000	9.000	14.000	0.900	0.930	0.710	0.165	0.260	0.990	0.340	4.400	41.000
June 28	1.000	156.900	60.000	100.000	2.410	1.330	1.570	0.395	0.590	1.140	0.260	3.600	11.000
June 28	2.000	139.100	53.000	77.000	2.150	2.630	1.500	0.294	0.610	1.220	0.317	3.700	23.00
June 28	3.00	754.800	235.000	213.000	2.620	1.510	3.910	1.075	1.610	2.510	0.150	3.100	37.000
June 29	1.000	30.300	15.000	26.000	0.000	0.310	0.300	0.021	0.000	1.150	0.250	4.100	15.000
June 29	2.000	166.600	131.000	106.000	6.690	0.970	1.060	0.344	0.470	1.380	0.220	3.400	18.000
June 29	3.000	47.200	51.000	63.000	0.250	0.630	0.690	0.159	0.200	1.320	0.330	3.600	27.000

Concentrations are given in $\mu\text{g Cl}^{-1}$. Also shown are the liquid water content of the cloud over the sample period (LWC) in g m^{-3} , the pH of the cloud water sample taken immediately after collection, and the UV radiation flux taken over the sampling period (W m^{-2}). Average uncertainty in the carboxylic acid concentration was $9 \pm 9\%$ with a maximum of 36%. The uncertainties in the inorganic concentrations were 21% or less. The pH measurements are accurate to ± 0.1 pH unit.

Table 2
Air equivalent concentrations of the species analyzed in the cloud water samples

Date (1999)	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Acetate	Formate	Oxalate	Glutarate	Succinate	Malonate
June 21	3.24	0.1000	0.3700	0.0330	0.0320	0.0080	0.0020	0.0000	0.0050
June 21	1.26	0.0800	0.3500	0.0000	0.0080	0.0080	0.0000	0.0000	0.0040
June 21	1.73	0.1000	0.3900	0.0020	0.0100	0.0100	0.0000	0.0000	0.0060
June 24	1.23	0.0900	0.3300	0.0210	0.0200	0.0070	0.0010	0.0000	0.0160
June 24	0.60	0.2400	0.4400	0.0170	0.0170	0.0110	0.0030	0.0050	0.0220
June 24	0.92	0.3100	0.4800	0.0310	0.0320	0.0240	0.0060	0.0090	0.0340
June 28	4.08	1.5600	2.6000	0.0630	0.0350	0.0410	0.0100	0.0150	0.0300
June 28	4.41	1.6800	2.4400	0.0680	0.0830	0.0480	0.0090	0.0190	0.0390
June 28	11.32	3.5300	3.2000	0.0390	0.0230	0.0590	0.0160	0.0240	0.0380
June 29	0.76	0.3800	0.6500	0.0000	0.0080	0.0080	0.0010	0.0000	0.0290
June 29	3.67	2.8800	2.3300	0.1470	0.0210	0.0230	0.0080	0.0100	0.0300
June 29	1.56	1.6800	2.0800	0.0080	0.0210	0.0230	0.0050	0.0070	0.0440

Concentrations are in $\mu\text{g m}^{-3}$.

~ 4% of the total acidity. The residual acidity in all instances is, of course, associated with nitrate and NSS sulfate.

By taking the product (appropriately scaled) of the cloud water concentration and the cloud liquid water content, we can derive the concentration of the various chemical species per unit volume of air. While, technically, this procedure would yield only the component of each species partitioned into the aqueous phase, the data of Limbeck and Puxbaum (2000) suggest that around 90% of the four dicarboxylic acids we have measured would be scavenged into the cloud water. Hence, the air equivalents we calculate should be reasonably good approximations of ambient air concentrations of the four acids. These concentrations are shown in Table 2. Only five aerosol, clear-air filter samples are available for comparison. Furthermore, due to the low sample volumes, the only organic anion found at significant levels on the filters was oxalate, with a mean concentration (based on four samples) of $0.027 \mu\text{g m}^{-3}$. This compares quite well with the cloud water air equivalents mean of $0.024 \mu\text{g m}^{-3}$. Given the very small number of aerosol samples, however, a more telling comparison is that between the air equivalent cloud water concentrations and aerosol dicarboxylic acid concentrations reported by Kawamura and Sakaguchi over the Pacific Ocean. Average concentrations of oxalic, malonic, succinic and glutaric acid averaged 40, 11, 3, and 0.6 ng m^{-3} , respectively. These compare with the average corresponding concentration from Table 2 of 24, 27, 8 and 5 ng m^{-3} . While these means differ somewhat between the two data sets and the C_2/C_3 ratio in particular is substantially lower in this data set than in that of Kawamura and Sakaguchi (0.9 versus 3.6) or in the precipitation data of Sempere and Kawamura (1996), none of the species means significantly differ between the data sets.

4. Discussion and conclusions

The above data presentation, coupled with past work, evokes the question of the origin of the diacids in our venue. Kawamura and Sakaguchi (1999) suggested long-range transport

of pollutants from the Asian continent as an ultimate origin, at least of precursors, but with an in situ, photochemical mechanism(s) as the proximate source of oxalic and malonic acid (see also Satsumabayashi et al., 1990). To examine this issue, we perform a factor analysis (principle component) of our cloud water data set as given in Table 1. The component matrix (with varimax rotation) from this analysis is shown in Table 3. The mutual loading of the dicarboxylic acids with the inorganics and with acidity in the first component is quite interesting and suggests a common origin either in pollution or in sea salt or both. The second component is most heavily loaded by formate followed rather faintly by acetate. Certainly this suggests a separate source for at least formate, presumably in situ photochemistry. The two remaining factors are essentially loaded by LWC and ultraviolet radiation, respectively, neither of which appears to strongly covary with any of the chemical species.

Finally, there is the issue of the contribution of the dicarboxylic acids to the total air-equivalent mass. These acids in fact constitute, on average, $\sim 1.5\%$ of the total resolved mass and at most 4%. This contribution is consistent with the relative abundance found by Kawamura and Usukura (1993) in the western Pacific near Japan but appreciably higher than values found by Kawamura and Sakaguchi (1999) for the central Pacific, again suggesting a continental source for either the acids themselves or their precursors.

It is also of interest to analyze the potential impact of the measured concentrations of dicarboxylic acids on cloud drop activation. The masses just discussed are obviously far too small relative to that of the inorganic solutes to have any impact on the shape of the Köhler growth curve. However, as several previous studies have noted, relatively minute amounts of organics could significantly impact droplet surface tension and thus drop activation (e.g., Chuang et al., 1997). To examine this issue, we must estimate the concentration of our measured dicarboxylic acids when the drops containing them were near their critical radii. This has been done by comparing the measured mean cloud drop size during cloud water

Table 3
Rotated component matrix^a for the factor analysis

	Component			
	1	2	3	4
SO ₄ ²⁻	0.959	0.207	0.129	4.759e-02
NO ₃ ⁻	0.957	0.155	0.158	5.973e-02
NSS SO ₄ ²⁻	0.945	0.164	-0.136	-0.162
Malonate	0.927	-1.403e-02	-0.155	0.117
Succinate	0.918	0.267	8.732e-02	0.231
Glutarate	0.908	0.266	0.190	0.228
pH	-0.902	7.732e-02	0.357	0.160
Oxalate	0.885	0.249	0.199	0.289
Cl ⁻	0.770	0.253	0.470	0.316
Acetate	0.506	0.500	1.119e-02	-0.488
Formate	0.141	0.948	-3.574e-02	0.106
LWC	-1.506e-02	5.431e-02	-0.980	0.114
UV	0.234	0.101	-9.265e-02	0.878

Extraction method: principal component analysis.

Rotation method: varimax with kaiser normalization.

^a Rotation converged in seven iterations.

collection with estimates of the critical radii derived from the average solute mass per particle (taken as the bulk mass as given in Table 2 divided by the number of activated particles, i.e., cloud drop number concentration) and the application of Köhler theory. Dilution factors were calculated from the comparison of critical radii with measured cloud drop radii and applied (inversely) to the measured dicarboxylic concentration to arrive at those present when the droplets were near their critical radius. The data and derived values involved in this analysis are shown in Table 4. The concentrations of diacids are found to be less than or equal to 0.02 M near the droplet critical radii. From Shulman et al. (1996), diacid concentrations for any of the acids found in our samples would have to be at least 0.1 M or greater to exert any appreciable effect on droplet surface tension and thus droplet activation. Of course, other, unanalyzed dicarboxylic acids could be present which would also contribute to surface tension reduction. However, from the work of Kawamura and Sakaguchi (1999), we would not expect the total dicarboxylic acid concentration (exclusive of oxalic acid which contributes negligibly to surface tension reduction) to exceed twice that of the species analyzed here. Hence, the dicarboxylic acid concentrations plausibly present in our CCN near their critical radius are still too low to impact CCN activation.

This result stands in contrast to the study of Facchini et al. (1999) who found significant surface tension reduction by direct measurement in droplets derived from fogs in the Po Valley of Italy and vacuum evaporated down close to their critical radii. However, fogs in the Po Valley are notoriously polluted, with very high concentrations of organic surfactants not found in more remote environments. For this polluted venue, aerosol and cloud drop number concentrations are sufficiently high (cf., Noone et al., 1992) that the clouds are really not susceptible to albedo modulation through marginal changes in cloud drop number concentration. For the more plausible venue for indirect forcing explored here, the measured organic acid concentrations are not sufficiently high to produce a significant

Table 4

Data and derived values for analysis of dicarboxylic acid concentrations present in haze droplets near their critical radius for activation

Date	Case	CDNC (cm^{-3})	LWC (g m^{-3})	\bar{r}_{CDNC} (μm)	r_{crit} (μm)	Dilution	[Di Acids] r_{crit} (M) ^a
June 21	1	47	0.16	9	0.32	2.2×10^4	0.008
June 21	2	48	0.16	9	0.37	1.44×10^4	0.004
June 21	3	49	0.17	9	0.36	1.56×10^4	0.005
June 24	1	55	0.22	10	0.36	2.14×10^4	0.02
June 24	2	91	0.17	8	0.35	1.2×10^4	0.02
June 24	3	60	0.34	11	0.50	1.1×10^4	0.01
June 28	1	113	0.26	8	0.90	7.0×10^2	0.001
June 28	2	93	0.25	9	0.90	10^3	
June 28	3	71	0.15	8	0.9	7.0×10^2	0.003
June 29	1	93	0.25	9	0.45	8×10^3	0.02
June 29	2	107	0.22	8	0.9	7.0×10^2	0.001
June 29	3	108	0.33	9	0.9	10^3	0.002

^a Concentrations of malonic, succinic and glutaric acids; oxalic is not included since it has a negligible impact on surface tension at the concentrations encountered here (Shulman et al., 1996).

effect. Nevertheless, this preliminary analysis certainly does not preclude a modulation of cloud albedo by organics. A more complete chemical analysis of many more cloud water samples will be necessary before definitive conclusions can be reached.

Acknowledgements

This research was supported by grants from the Office of Naval Research (N00014-97-1-0132) and the National Science Foundation (ATM-9908411). We thank two anonymous reviewers for useful comments.

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