RAINWATER COMPOSITION IN ATHENS, GREECE

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Abstract—Wet precipitation-only samplers were used to collect wet deposition at two sites in the Athens basin, Greece for the period March 1986–February 1987.

Concentrations of major cations (H^+, NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+}) and major anions (Cl^-, NO_3^- and SO_4^{2-}) were determined for the first time in rainwater samples in Greece. Bicarbonate concentrations were calculated. The relative importance of natural and anthropogenic sources were estimated by a chemical balance. The majority of rain collected has a neutral or alkaline character. Acidity was due to the presence of H_2SO_4 and HNO_3. The statistical analysis of the correlation between the concentration of chemical species confirm the influence of natural and anthropogenic sources. In all samples, SO_4^{2-} concentrations exceed NO_3^- concentrations despite the dominance of low S oil burning in the region. The wet flux of S was calculated to be $0.34\ \mathrm{g\,m^{-2}\,a^{-1}}$.

Key word index: Acid rain, wet deposition, acid precipitation, sampling and analysis of rain, atmospheric acidity, precipitation chemistry.

1. INTRODUCTION

Acid precipitation, composed primarily of dilute H_2SO_4 and HNO_3 and originating from the burning of fossil fuels, has become one of the major environmental problems and has been the subject of intense research in western Europe and northeastern U.S. for about 20 years (Pierson and Chang, 1986; EEC, 1985; O'Sullivan, 1985; Environ. Res., 1983; Hansen and Hidy, 1982; Beilke, 1983).

Acid rain is blamed for tree damage, acidification of water lakes and deterioration of marble structures and ancient monuments (O'Sullivan, 1985; Cheng *et al.*, 1987; Vocom, 1979; Skoulikidis, 1983). In recent years, special attention has been drawn to the deterioration of marbles of Acropolis (Parthenon) in Athens, Greece (Vocom, 1979; Skoulikidis, 1983; EEC, 1985).

The phenomenon of acid deposition in Athens, Greece, to the best of our knowledge, has not been investigated by direct measurement of samples of acid rain and very little precipitation data can be found in the literature (O'Sullivan, 1985; Samara *et al.*, 1987; Howells and Kallend, 1984).

The greater Athens area, a basin of approx. $450\ \mathrm{km^2}$, has grown rapidly in recent decades by population shifts and increased overall industrialization. As a consequence, pollution has reached high levels and could be a hazard to humans and ancient monuments (Lalas *et al.*, 1982; EPCPA, Terminal Report, 1982; OECD, 1983; Lekakis, 1984).

This paper presents results for precipitation chemistry (14 parameters) at two locations in Athens for the period March 1986–February 1987 and discusses these results in terms of chemical source balance and equilibrium models.

2. STATION NETWORK, SAMPLING AND ANALYSIS

The choice of station network and location of sampling equipment was made according to internationally accepted specifications (Still, 1983). Sampling devices were installed at two different sites: site A: University campus at the limits of urban area and site B: Observatory hill in the centre of Athens, near Acropolis area. Figure 1 shows the locations of sample collection sites. The samplers at site A and B were placed 10 and 4 m above ground level, respectively.

Samples were collected using AC or battery operated, home made, automatic wet-only samplers. The sampler collects samples of wet deposition in a cylindrical polyethylene bucket that is uncovered automatically by means of the conductivity type precipitation sensor, at the inception of the first drops and covered immediately after the end of rainfall. Dry deposition is effectively excluded in this manner. The polyethylene bucket with 30 cm diameter (opening area $706\ \mathrm{cm^2}$), was cleaned with distilled water and laboratory paper, immediately after sample collection. During dry periods the cleaning of the bucket was frequent enough to avoid dry deposition and contamination by local dust.

Samples were collected over 12 h periods and labeled as D (daily events ending at 20:00) and as N (night events ending at 08:00). The sample period extended from March 1986 to February 1987 and a

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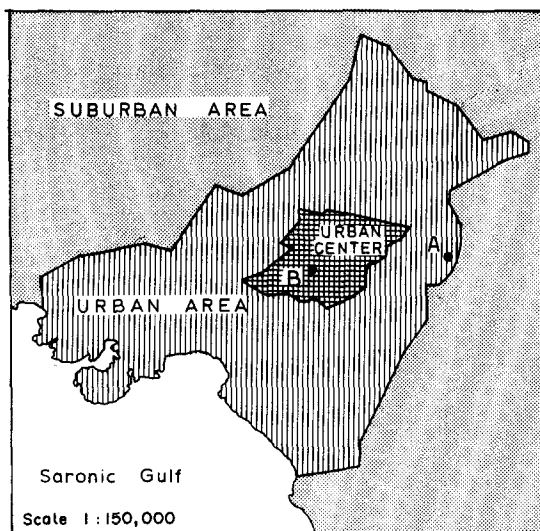‡ Author to whom correspondence should be addressed.

Fig. 1. Locations of sample collection sites. The urban center is an area of about 9000 hectares in which vehicles are allowed to travel only on alternative days. A: Sampling site at the limits of the urban area; B: sampling site in the center of Athens, near Acropolis.

the collectors. Many of the samples were also titrated coulometrically for the determination of rain water total acidity (Liberti et al., 1972). These samples were purged for 10 min with nitrogen gas to remove CO_2. A 50.00 ml aliquot of sample, the same used for pH measurement, was titrated in a nitrogen atmosphere with the generated OH^-. The end points of titration curves were calculated using Gran's plots. Chemical analysis was based on methods recommended by international organizations (APWA, 1981; EPA, 1979; Norwegian Institute for Air Research, 1977). Concentrations of SO_4^{2-}, NO_3^-, Cl^- and NH_4^+ were measured photometrically. Flame photometry was used for determination of K^+ and Na^+, while atomic absorption spectrophotometry was used for determination of Ca^{2+} and Mg^{2+}. Fluorides and organic anions ($HCOO^-$, CH_3COO^-) were not determined. The minimum rain volume needed for a complete analysis for the determination of the above mentioned species will be 150 ml and this corresponds on our rain collector to 2.5 mm rain amount. Rain samples of smaller volumes can be analyzed, after measuring in the initial sample the pH, conductivity and acidity, by diluting the rain sample with distilled water to the appropriate volume.

total number of 74 samples were collected during this period. All rainfall was collected. There were no rain events during the summer period, i.e. July, August and September. Rain samples were stored in pre-cleaned polyethylene bottles (Moody and Lindstrom, 1977) and sent immediately to the laboratory for chemical analysis.

Rainfall height and quantity was roughly estimated by measuring the volume of collected sample in a graduated cylinder. Upon receipt in the laboratory, samples were filtered out. The insoluble fraction was dried for 1 h at 105°C, weighed and stored. The filtrate was stored at 4°C in the refrigerator. An aliquot was stored separately after the addition of some $CHCl_3$ drops in order to avoid any bacterial action. This aliquot was used for the determination of NH_4^+ and NO_3^-.

Conductivity measurements were performed using a L 21. L Pusl model conductivity meter. The conductivity cell was calibrated against standard KCl solutions.

Measurement of sample pH was made without stirring using a Corning Model 12 pH-meter equipment with a combination glass electrode. The pH-meter was calibrated before every measurement, using standard buffer solutions at pH 4.00 and 7.00. The low ionic strength of rain samples causes instability problems in reading of pH values. The addition of KCl into the samples, in order to correct the ionic strength, is not taken into account in the reported pH values. pH measurements cannot be reported with a greater confidence than 0.1 pH unit (Koch and Marinenko, 1983). These measurements were typically completed during the same day that samples were removed from

3. RESULTS AND DISCUSSION

Mean concentrations and rainfall pattern

The precipitation monthly weighted mean concentrations during the period March 1986–February 1987 are given in Table 1.

Quality assurance of chemical analysis

Weighted mean concentrations reflect the analytical criteria imposed by individual samples. Results of samples were checked for internal consistency with charge or conductivity balance to ensure that all major cation and anion concentrations were accurately determined.

The charge balance of cation and anion equivalent concentration is a necessary but not sufficient condition that the major ionic species have been determined. Linear correlation between measured anion and cation equivalent concentration was significant at the 95% confidence level ($r = 0.949$, $n = 46$). Σ^- and Σ^+ values are presented in Table 2 (Grundahl and Keiding, 1985).

The agreement between measured conductivity and specific conductivity calculated from ionic conductances and measured ionic concentrations is a necessary and sufficient condition that the major ionic species concentrations have been determined. The linear correlation coefficient (r) between measured and calculated conductivities was 0.984 for $n = 31$.

Equilibrium model for bicarbonate in rain

The sum of anionic concentrations $\Sigma^- = ([Cl^-] + [NO_3^-] + 2[SO_4^{2-}] + [HCO_3^-])$ and the sum

Table 1. Monthly volume weighted mean chemical concentration and standard deviation in $\mu eq\ \ell^{-1}$ for rain water samples collected in Athens, Greece

Month	03/86	04/86	05/86	06/86	10/86	11/86	12/86	01/87	02/87	Year mean ±SD
Total number of rain samples*	4	8	9	10	17	6	8	6	6	74
Total rain amount (mm)	19.5	13.4	43.8	78.4	74.3	41.2	42.4	27.3	36.2	377
Conductivity (μS cm^{-1})	12.6±13.9	36.1±21.6	29.5±12.7	20.2±24.8	14.8±22.0	15.8±10.8	28.6±53.0	246±214	29.4±28.8	39.0±55.5
[IPM] (mg ℓ^{-1}) ●	15.3±22.0	85.4±356	9.2±31.6	17.6±452	18.0±968	8.3±11.2	26.2±353	170±225	—	30.5±520
[H$^+$]$_f$	2.7±1.7	0.2±0.1	23.7±21.0	0.9±0.6	0.6±1.2	0.4±0.10	0.2±0.1	0.6±1.8	0.1±0.0	4.00±14.2
[Na$^+$]	37.1±47.8	44.3±92.7	50.1±71.1	26.3±16.7	61.4±202	46.6±19.3	89.3±80.4	231±99.7	128±50.8	67.7±116
[K$^+$]	19.7±7.1	31.6±140	15.3±7.3	10.9±135	18.0±91.0	15.0±2.3	4.9±40.3	27.2±22.0	7.4±4.7	14.5±82.8
[Ca^{2+}]	114±185	291±97.6	67.8±33.8	94.3±128	74.0±131	74.0±60.0	122±643	461±203	285±240	137±281
[Mg^{2+}]	21.2±23.7	58.2±332	16.6±37.1	15.4±199	14.4±17.2	12.0±7.4	28.2±144	124±95.4	41.8±11.6	31.0±155
[NH$_4^+$]	14.9±6.8	12.3±8.6	39.7±92.7	24.0±48.5	16.4±200	22.1±15.3	16.8±73.5	19.2±8.4	21.6±8.5	21.9±108
[Cl$^-$]	56.8±39.4	137±575	75.1±117	74.4±216	47.0±1306	37.9±26.1	97.1±160	333±185	139±34.7	94.2±680
[NO$_3^-$]	34.9±64.8	30.8±289	31.1±30.7	28.8±108	17.5±484	15.3±9.6	17.2±130	30.8±9.3	18.5±10.6	23.6±260
[SO$_4^{2-}$]$_t$	133±46.4	182±31.2	157±47.1	46.0±54.3	54.6±3244	65.2±60.2	89.9±108	244±117	146±119	100±1562
[HCO$_3^-$]●,†	29.9±80.4	124±32.9	5.2±3.8	45.3±97.1	41.6±54.2	46.6±36.7	78.4±140	246±128	174±121	72.4±97.4
[SO$_4^{2-}$]/[NO$_3^-$]†	11.0	8.9	5.5	1.6	3.6	4.2	8.2	9.0	7.8	5.4
[Na$^+$]/[Cl$^-$]†	0.62	0.37	0.77	0.37	0.96	1.40	0.91	0.91	0.89	0.81
Σ$^-$/Σ$^+$‡	1.15	0.95	1.17	1.04	0.81	1.04	1.07	0.95	1.01	1.02

Σ$^+$: is the sum of cationic concentrations, Σ$^-$: is the sum of anionic concentrations, Σ$^-$/Σ$^+$: is the arithmetic mean value, ●: calculated. $\pm$[H$^+$]$_f = 10^{-pH}$.
* For two samplers (two sites).
† Selected values.
IPM = Insoluble particulate matter. [SO$_4^{2-}$]$_t$ = total sulfates from marine and terrestrial sources.
— = Not measured.

Table 2. pH values, total acidity concentrations and anion–cation charge balances of selected samples out of 50 collected rain samples

pH	Total acidity (μeq ℓ^{-1})	Σ^* (μeq ℓ^{-1})	Σ^+ (μeq ℓ^{-1})	Σ^-/Σ^+
–	–	109	91	1.20
–	155	679	631	1.08
4.20	–	321	265	1.21
4.30	154	286	288	0.99
4.60	101	–	–	–
5.40	–	170	116	1.47
5.40	59.5	115	169	0.68
5.40	77.0	293	175	1.68
5.50	–	186	120	1.54
5.60	67.4	189	143	1.33
5.80	73.3	92	82	1.13
5.90	–	379	400	0.95
6.00	78.9	171	136	1.26
6.20	54.8	51	70	0.72
6.30	62.6	346	355	0.97
6.30	55.1	122	163	0.73
6.30	13.4	131	155	0.85
6.40	58.9	–	–	–
6.40	–	200	148	1.35
6.50	23.8	161	205	0.79
6.50	11.4	131	144	0.91
6.50	43.0	133	148	0.90
6.50	–	167	135	1.23
6.60	–	421	441	0.95
6.60	35.9	358	659	0.54
6.60	–	636	661	0.96
6.70	37.4	257	392	0.66
6.70	–	554	462	1.20
6.70	–	221	245	0.91
6.80	–	394	449	0.88
6.90	11.5	576	647	0.89
6.90	–	254	229	1.11
7.00	36.3	–	–	–
7.00	–	718	952	0.75
7.00	–	293	243	1.21
7.00	–	281	259	1.08
7.00	–	319	358	0.89
7.10	27.2	277	282	0.98
7.10	14.8	169	176	0.96
7.10	26.2	291	311	0.94
7.10	–	231	233	0.99
7.20	–	436	463	0.96
7.20	44.1	166	137	1.21
7.30	–	767	845	0.91
7.40	–	431	473	0.91
7.40	–	463	525	0.88
7.70	–	–	–	–
7.80	–	1524	1325	1.15
7.90	–	593	657	0.90
8.10	13.4	406	300	1.35

Σ^- and Σ^+: Sum of the anion and cation concentrations, respectively in μeq ℓ^{-1}.

– = Not measured and therefore not calculated.

*The Σ^- values were calculated taking into account the [HCO_3^-] for samples with pH $\geqslant$ 5.6.

of cationic concentrations $\Sigma^+ = ([H^+] + [NH_4^+] + [Na^+] + [K^+] + 2[Mg^{2+}] + 2[Ca^{2+}])$ was calculated for 46 samples. The Σ^-/Σ^+ ratio was systematically < 1 due to the loss of CO_2 by buffering of

atmospheric acids by $CaCO_3$ whereas the ratio was increased to about 1 when the [HCO_3^-] were taken into calculation. For samples with pH $\leqslant$ 5.6 the concentration of [HCO_3^-] was considered negligible. Samples with pH > 5.6 have high concentrations of Ca^{2+} and Mg^{2+}, which can be attributed to neutralization process of rain acids by suspended $CaCO_3$ and $MgCO_3$ in air originated mostly from roadside dust. For these samples the bicarbonate concentration was calculated using Equation (1).

$$[HCO_3^-] = \{K(S + [Me^{2+}])\}/(10^{-pH} + K) \quad (1)$$

where: $K = 4.2 \times 10^{-7}$, first ionization constant of H_2CO_3, $S = 3.05 \times 10^{-2}$ mol ℓ^{-1}, solubility of CO_2 in water at 25°C and 1 atm, $[Me^{2+}] = ([Ca^{2+}] + [Mg^{2+}])$ mol ℓ^{-1}. The correlation coefficient between Σ^- and Σ^+ was calculated to be 0.949 ($n = 46$).

Significance of pH and acidity variations

Figure 2 illustrates the pH distribution (from 4.2 to 8.1) of 62 samples over the period March 1986–February 1987. About 20% of rain samples appeared with pH $\leqslant$ 5.6 and only 5% of samples with pH < 4.5, which can be considered as the lower pH of natural rainwater (Charlson and Rodhe, 1982). Samples with total amount of rain greater than 15 mm show pH values very close to 5.6. Acidic pH values reveal the presence of strong acids in rain water, while neutral or alkaline pH values are attributed to neutralized acids by ammonia, sea water salt and soil dust (predominantly $CaCO_3$ and $MgCO_3$). It is known that in Athens area limestone is a predominant constituent in soil and airborne dust (EPCPA, 1982). High concentrations of solid (undissolved) materials in samples of
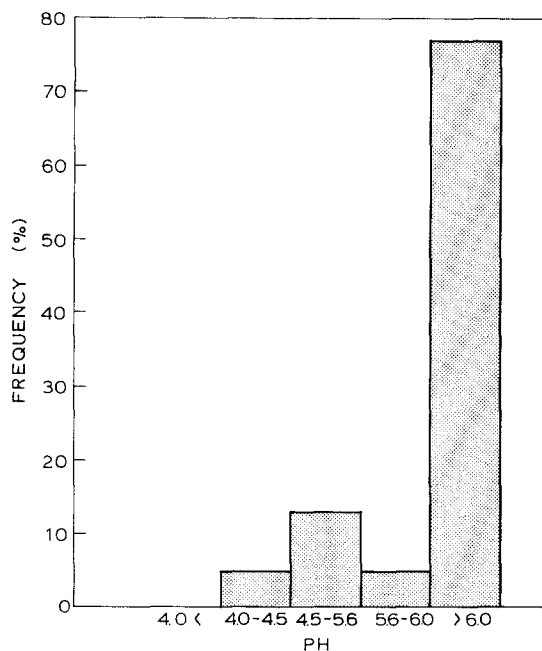

Fig. 2. pH distribution.

small amount of rain are responsible for the neutral pH values.

The comparison of pH and total acidity values for 25 selected samples is summarized in Table 2. There is a good correlation ($r = 0.783$ for $n = 25$) between total acidity concentrations and free acidity ($[H^+]$ $= 10^{-pH}$) values. Total acidity concentrations vary from zero up to 155 $\mu eq\,\ell^{-1}$, while 18 $\mu eq\,\ell^{-1}$ is the acidity for the uncontaminated rain (Zajicek, 1985). The increase of weak acid concentration in a city–industrial area was earlier reported by Kasina (1980).

Determination of chemical sources

Natural or anthropogenic sources and processes (sea salt, soil dust, HNO_3, H_2SO_4, NH_3) are responsible for the ionic profile of rainwater. The ionic concentrations reported here are without any correction for the contribution of sources.

The $[Na^+]/[Cl^-]$ ratio year weighted mean value for $n = 57$ rain samples was found to be 0.81, while Grundahl et al. (1985) reported a value of 0.82. The corresponding value for seawater is 0.86 (Brewer, 1975). This finding means that rainwater composition is mostly influenced by marine sprays coming by Saronicos gulf, when winds are favorable (Liljestrand and Morgan, 1981).

Strong acids (H_2SO_4, HNO_3) of anthropogenic origin are responsible for acid rain incidents. There is poor correlation ($r = 0.29$) between hydronium concentration $[H^+]_f$ and $[SO_4^{2-}]_t$ and an even worse correlation ($r = 0.08$) between $[H^+]_f$ and $[NO_3^-]$. There is a qualitative agreement with correlation values reported by Gorham et al. (1984).

Neutral to alkaline pH values indicate that neutralization takes place as a major process in rain water. In order to identify the ions which partake of the phenomenon of rain acidification and to quantify their probability of participation, the chemical concentrations were analyzed for linear correlation. Table 3 gives the values of the correlation coefficients for selected samples collected at the level of 95%. From these results the following is shown.

There is a remarkable correlation between the undissolved solids and the most of chemical species ($[NO_3^-]$, $[Cl^-]$, $[NH_4^+]$, $[SO_4^{2-}]$, $[K^+]$, $[HCO_3^-]$). The chemical profile of rainwater in Athens seems to be very influenced by the presence of electrolytes originated from neutralization processes with sea-salts and soil-dust. Nitrates and sulfates are probably produced by ionization of relative electrolytes in water phase. This statement is supported by the relatively neutral to alkaline pH values and the poor correlation between free acidity—$[SO_4^{2-}]_t$, $[NO_3^-]$ reported. $[NH_4^+]$ ions are correlated with $[Cl^-]$ (0.89), $[NO_3^-]$ (0.94), $[SO_4^{2-}]_t$ (0.90), and solids (0.89) indicating the participation of sea water and soil in the neutralization process.

Finally it can be noted that a group of strongly correlated elements Cl^-, Na^+, K^+ and Mg^{2+}, represents the maritime source.

In conclusion, the same chemical pattern was observed in acid rain collected in Spain (Lacaux et al., 1987), which confirms the anthropogenic and natural origin of elements.

Sulfates in rain water

Acidity was due mainly to the presence of H_2SO_4 and HNO_3 and perhaps to HCl. The ratio of ionic $[SO_4^{2-}]/[NO_3^-]$ was typically >5, i.e. the weighted year mean ratio was calculated to be 5.4. The wet flux of S was calculated to be 0.34 $g\,m^{-2}\,a^{-1}$.

4. CONCLUSIONS

The major components causing the net precipitation acidity in Athens, Greece are pollutant H_2SO_4 and HNO_3. These acids are partially neutralized by gaseous NH_3 and alkaline soil dust and marine sprays in the atmosphere.

The majority of rain collected has a neutral or alkaline character. Indeed only 20% have a pH $\leqslant 5.6$.

The statistical analysis of the correlations between the concentrations of chemical species confirm the anthropogenic character of the elements.

Table 3. Correlation coefficients at the probability of 95% for all samples (March 1986–February 1987)

	$[H^+]_f$	$[Na^+]$	$[K^+]$	$[Ca^{2+}]$	$[Mg^{2+}]$	$[NH_4^+]$	$[Cl^-]$	$[NO_3^-]$	$[SO_4^{2-}]_t$	$[HCO_3^-]$	$[IPM]$
$[H^+]_f$	1.00										
$[Na^+]$	−0.18	1.00									
$[K^+]$	−0.08	0.79	1.00								
$[Ca^{2+}]$	−0.16	0.44	0.23	1.00							
$[Mg^{2+}]$	−0.08	0.34	0.91	0.59	1.00						
$[NH_4^+]$	0.46	0.25	0.29	0.30	0.17	1.00					
$[Cl^-]$	−0.03	0.84	0.85	0.19	0.32	0.89	1.00				
$[NO_3^-]$	0.08	0.26	0.59	0.34	0.33	0.94	0.94	1.00			
$[SO_4^{2-}]_t$	0.29	0.30	0.09	0.15	0.00	0.90	0.90	0.90	1.00		
$[HCO_3^-]$	−0.30	0.76	0.13	0.96	0.86	0.41	0.80	0.56	0.66	1.00	
$[IPM]$	−0.09	0.27	0.69	0.33	0.34	0.89	0.92	0.93	0.88	0.63	1.00

$[H^+]_f$ = Free acidity ($[H^+] = 10^{-pH}$).
$[SO_4^{2-}]_t$ = Total sulfates from marine and terrestrial sources.
$[IPM]$ = Insoluble particulate matter (undissolved solids).

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