Effects of Acid Deposition on Dissolution of Carbonate Stone During Summer Storms in the Adirondack Mountains, New York, 1987-89

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FIGURES

3-6. Graphs showing:

TABLES

CONVERSION FACTORS AND ABBREVIATIONS

Multiply	By	To obtain
Calcium ion, milliequivalent per liter (meq/L)	20.04	part per million Ca^{+2}
Hydrogen ion, milliequivalent per liter (meq/L)	1.0079	part per million H^{+}
kilometer (km)	0.6214	mile
liter (L)	1.057	quart
meter (m)	3.281	foot
millimeter (mm)	3.937×10^{-2}	inch
microgram (µg)	3.527×10^{-8}	ounce, avoirdupois
micrometer (µm)	3.937×10^{-5}	inch
milliliter (mL)	0.03382	ounce, fluid
Sulfate ion, milliequivalent per liter (meq/L)	48.0288	part per million SO_4^{2-}

Degree Celsius (°C) may be converted to degree Fahrenheit (°F) by using the following equation:

$$°F = 9/5\ (°C) + 32.$$

The following terms and abbreviations also are used in this report:

millimeter per hour (mm/hr)

milliequivalent per square meter (meq/m^2)

microgram per square meter (µg/m^2)

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Abstract

This study is part of a long-term research program designed to identify and quantify acid rain damage to carbonate stone. Acidic deposition accelerates the dissolution of carbonate-stone monuments and building materials. Sequential sampling of runoff from carbonate-stone (marble) and glass (reference) microcatchments in the Adirondack Mountains in New York State provided a detailed record of the episodic fluctuations in rain rate and runoff chemistry during individual summer storms. Rain rate and chemical concentrations from carbonate-stone and glass runoff fluctuated three to tenfold during storms. Net calcium-ion concentrations from the carbonate-stone runoff, a measure of stone dissolution, typically fluctuated twofold during these storms. High net sulfate and net calcium concentrations in the first effective runoff at the start of a storm indicated that atmospheric pollutants deposited on the stone surface during dry periods formed calcium sulfate minerals, an important process in carbonate stone dissolution. Dissolution of the carbonate stone generally increased up to twofold during coincident episodes of low rain rate (less than 5 millimeters per hour) and decreased rainfall (glass runoff) pH (less than 4.0); episodes of high rain rate (cloudbursts) were coincident with a rapid increase in rainfall pH and also a rapid decrease in the dissolution of carbonate-stone. During a storm, it seems the most important factors causing increased dissolution of carbonate stone are coincident periods of low rain rate and decreased rainfall pH. Dissolution of the carbonate stone decreased slightly as the rain rate exceeded about 5 millimeters per hour, probably in response to rapidly increasing rainfall pH during episodes of high rain rate and shorter contact time between the runoff and the stone surface. High runoff rates resulting from cloudbursts remove calcium sulfate minerals formed during dry periods prior to storms and also remove dissolution products formed in large measure by chemical weathering as a result of episodes of low rain rate and decreased rainfall pH during a storm.

INTRODUCTION

Acidic deposition is recognized as an important cause of carbonate-stone dissolution in selected environments. Past studies (Johnsson and Reddy, 1990; Borchert, 1986; Reddy and others, 1985; Seymour and Stout, 1983; Kins, 1982; and Dawson, 1978) indicate rain rate and chemistry fluctuate during individual storms. There is little understanding, however, of how these fluctuations affect the dissolution of carbonate stone. Little research on carbonate-stone dissolution by acidic deposition has been done that involves the measurement of a complete suite of climatological, meteorological, and atmospheric pollutant factors during the sequential sampling of rainfall runoff from an individual storm. To gain a better understanding of the interaction of acidic deposition with carbonate-stone building materials, it is necessary to monitor the episodic fluctuations in rain rate and chemistry during individual storms.

Background

Acidic deposition, defined as wet deposition in the form of acid rain, acid snow, or acid fog, and the dry deposition of atmospheric pollutants, has been linked to the acidification of European and North American lakes (Cosby and others, 1985; Driscoll and Newton, 1985), to forest decline (Tomlinson, 1983), and to the accelerated dissolution and recession of monuments and carbonate-stone building materials (Baedecker and others, 1990; Lipfert, 1989; Reddy, 1988; Reddy and others, 1986b). Carbonate-stone building materials are subjected to changing natural factors such as temperature, wind, humidity, and precipitation that may contribute to the physical and chemical weathering and subsequent dissolution of the stone. Generally, this dissolution is a slow process; however, it is accelerated when the stone also is exposed to atmospheric pollutants in the presence of water (Baedecker, and others, 1990). Oxides of sulfur and nitrogen react in the atmosphere to produce sulfuric and nitric acids. These acids in rainfall react with the stone surface. Gaseous and particulate pollutants, in the form of dry deposition, also may react with the stone surface in the presence of moisture. Onsite field exposure studies (Reddy and others, 1989; Reddy and Werner, 1985; Jaynes and Cooke, 1987) indicate that stone damage (dissolution and associated surface recession) increased with decreased rain pH.

Stone-dissolution research is based on the net, long-term effect of atmospheric pollutants such as sulfur dioxide on carbonate-stone building materials and also of the chemistry of runoff from carbonate stone. Furthermore, acidity of rainfall and analyses of runoff usually are determined from integrated samples that represent average concentrations over a particular time period and do not describe the fluctuations of acidity of rainfall or chemistry of runoff from carbonate stone during individual storms. Although the integrated precipitation samples used in most acid rain studies can provide valuable information on precipitation chemistry, integrated samples may obscure detailed changes in rainfall and runoff chemistry during storms. Dissolution of carbonate-stone building materials by acidic deposition requires knowledge of rainfall and runoff characteristics as the storm progresses. Sequential sampling of runoff from individual storms could be critical in determining mechanisms responsible for the dissolution of carbonate stone due to acid deposition.

This study conducted by the U.S. Geological Survey was part of a long-term research program designed to identify and quantify acid rain damage to carbonate stone (Reddy, 1988). The analytical results presented in this report are intended to aid researchers in the assessment of the effects of acidic deposition on building materials. The objective of this study is to identify factors or a combination of factors that influence the dissolution of carbonate stone during individual storms.

Purpose and Scope

This report describes an onsite, prototype experimental system designed to monitor the dissolution of carbonate stone exposed to atmospheric pollutants and acid rain during seven selected summer storms during 1987–89. Results from onsite measurements and laboratory analyses of physical and chemical constituents of runoff collected from carbonate-stone (marble) and glass (reference) microcatchments were used to determine fluctuation of rainfall and runoff chemistry. Microcatchment is defined in this report as a device consisting of a fixed area and surface material that captures water used to measure atmospheric inputs and the quantity of material lost from a carbonate stone as a result of exposure to direct rainfall. Stone dissolution and acid loading to the stone surface during an individual storm were estimated from these results. Quantification of carbonate-stone dissolution during an individual storm was determined by measuring the quantity of calcium ions in runoff samples. Factors influencing dissolution quantification were determined by relating fluctuations in rainfall rate and chemistry to changes in runoff chemistry from the carbonate stone.

Description of the Study Site

The study site is located in Huntington Wildlife Forest at the Adirondack Ecological Center (a unit of the College of Environmental Science and Forestry, State University of New York) west of the town of Newcomb in the Central Adirondack Mountains of New York (fig. 1). The State University of New York has maintained a climatological and meteorological monitoring station at the site since 1940, and the New York State Department of Environmental Conservation (NYDEC) has monitored concentrations of sulfur dioxide, nitrogen oxides, and ozone at the site from the early 1980's until July 1989. The site is part of the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) and seems to be well-suited for studies of acid-rain effects. From 1984 to 1989, the mean volume-weighted pH of precipitation was 4.39 (NADP/NTN, written commun., 1991); concentrations of other anthropogenic pollutants generally were small. Because there are no nearby point sources of acid rain precursors, and traffic volume in the area is minimal, sources of acidic deposition to the site are believed to be regional.

Acknowledgments

The authors express their appreciation to Gwen Scott of the National Acid Precipitation Assessment Program/National Trends Network (NADP/NTN) for their cooperation and assistance. Special thanks to Charlotte Demers of Adirondack Ecological Center, State University of New York, for her assistance with onsite operations.

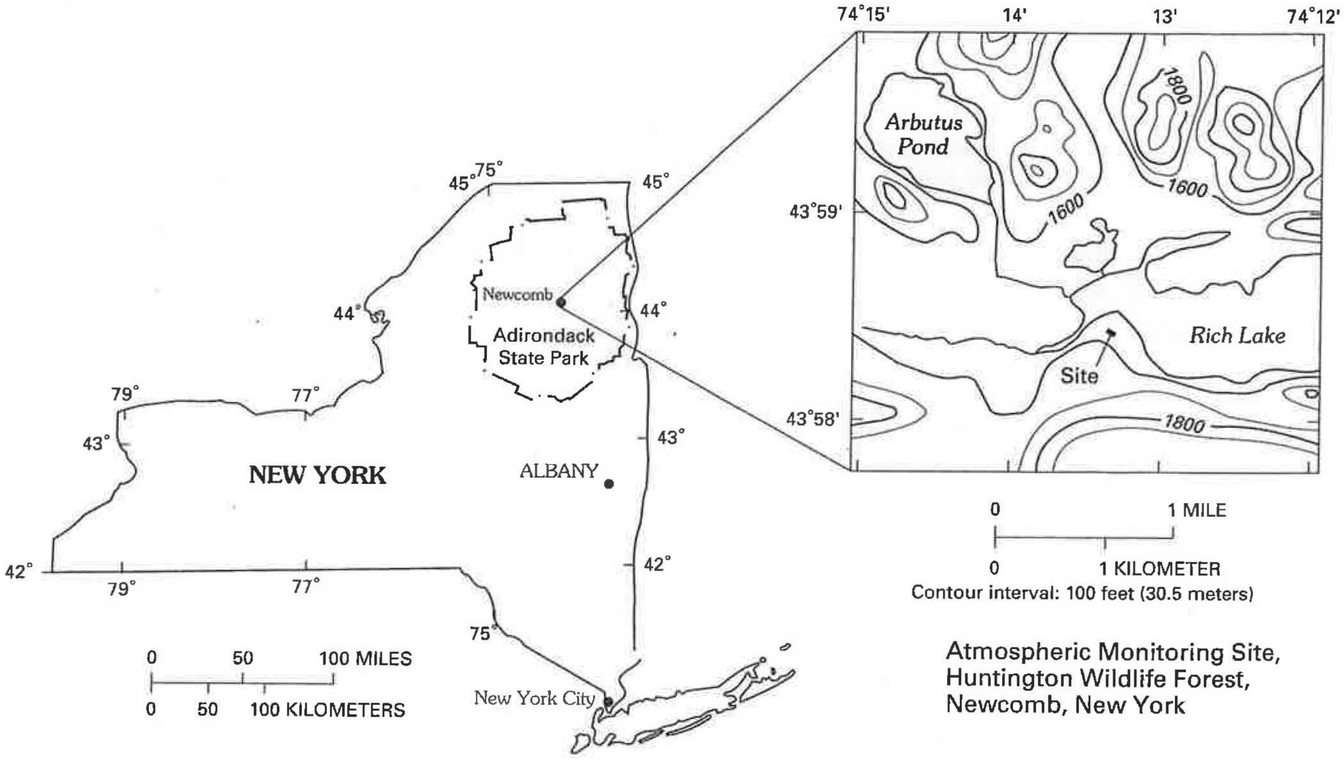

Figure 1. Location of the study site.

EXPERIMENTAL DESIGN

The onsite experimental system consisted of two microcatchments; a 0.1858 square meter (m^2) slab of Shelburne Marble, a stone commonly used as a building material and in monuments (Ross, 1985) and a Pyrex glass reference plate of the same size (sand blasted to simulate the roughness of the marble surface). A shield was placed over the lower end of the microcatchments covering 0.0697 m^2 of each surface to prevent sample-vial overflow of the runoff. The total exposed area of each surface was 0.1161 m^2. Exposure conditions of the glass and marble microcatchments were chosen to be the same as those for other materials-damage studies sponsored by the National Atmospheric Precipitation Assessment Program (NAPAP) (Flinn and others, 1986; Sherwood and Doe, 1984). The glass and marble microcatchments were tilted at 30° from the horizontal facing south, and the exposed section was uncovered at all times. Runoff was collected from the glass and marble microcatchments using a volume-based sequential sampler. For further description of the glass and marble microcatchments and their holding rack (Reddy, 1988; McGee, 1989). In addition, a suite of meteorological, climatological, and atmospheric-pollutant data were recorded continuously at the site. Table 1 describes and figure 2 shows the monitoring and sampling equipment employed in this study. Redundant sensors were placed at the site for quality assurance and alternates for possible sensor failure.

Sequential-Runoff Samplers and Tipping-Bucket Raingage

Two volume-based Gilson Escargot SC-30 sequential samplers were placed in a housing under the glass and marble microcatchments (1 in fig. 2). Runoff was directed to the escargot sampling tray with Tygon tubing. The sampler's circuitry was interfaced with a Weathertronics 6010 tipping-bucket raingage (2 in fig. 2). After every 0.254 mm of rainfall, the bucket tipped, and a mercury switch made a momentary closure, sending a voltage signal to the sequential samplers that advanced the escargot trays. Rainfall rate was calculated using data from the tipping-bucket raingage. The time of each tip was recorded on a CR-21 micrologger (Campbell Scientific). Each sequential sample represented 0.254 mm of rainfall. Three successive samples were aggregated to facilitate chemical analysis of the runoff. Therefore, each aggregated sequential sample represented 0.762 mm of rainfall. The rain rate during the time represented by the sequential sample was calculated by dividing 0.762 mm of rainfall by the total time elapsed during three tips of the raingage. Laquer (1990a) provides a thorough literature review of various sequential precipitation-sampling methods.

Table 1. Instrumentation used, factors measured, and measurement intervals at Newcomb, New York, during the sequential sampling of seven selected summer storms during 1987 through 1989

Instrumentation	Factors measured	Measurement interval
Sequential runoff samplers[1]	pH (onsite) Specific conductance (onsite) Major anions and cations (laboratory)	90 milliliter aliquots
Tipping-bucket raingage[2]	Volume of rain (advances the sequential sampler)	0.254 millimeter
U.S. Geological Survey meteorological station[3] (3-meter elevation)	Windspeed and direction Solar radiation Relative humidity Air temperature	15 minutes
Wetness sensors	Relative humidity[4] Leaf wetness (gypsum-coated circuit grid)[5] Stone wetness (limestone block resistor)	1 hour
Continuous precipitation monitor[6]	Air temperature pH Specific conductance Precipitation amount	1 minute

Instrumentation	Ancillary measurements	Measurement interval
NYDEC[7] air-quality monitors	Sulfur dioxide Nitrogen oxides Ozone	1 hour
NYDEC[7] meteorological station (3-meter elevation)	Windspeed and direction Relative humidity Air temperature	1 hour
HIF[8] meteorological station (30-meter elevation 0.8 kilometer north of site)	Windspeed and direction Relative humidity Leaf wetness Volume of rain Air temperature	1 hour

[1] See figure 2.

[2] See figure 2.

[3] See figure 2.

[4] See figure 2.

[5] See figure 2.

[6] See figure 2.

[7] New York Department of Environmental Protection.

[8] Huntington Forest Integrated Forest Study (State University of New York).

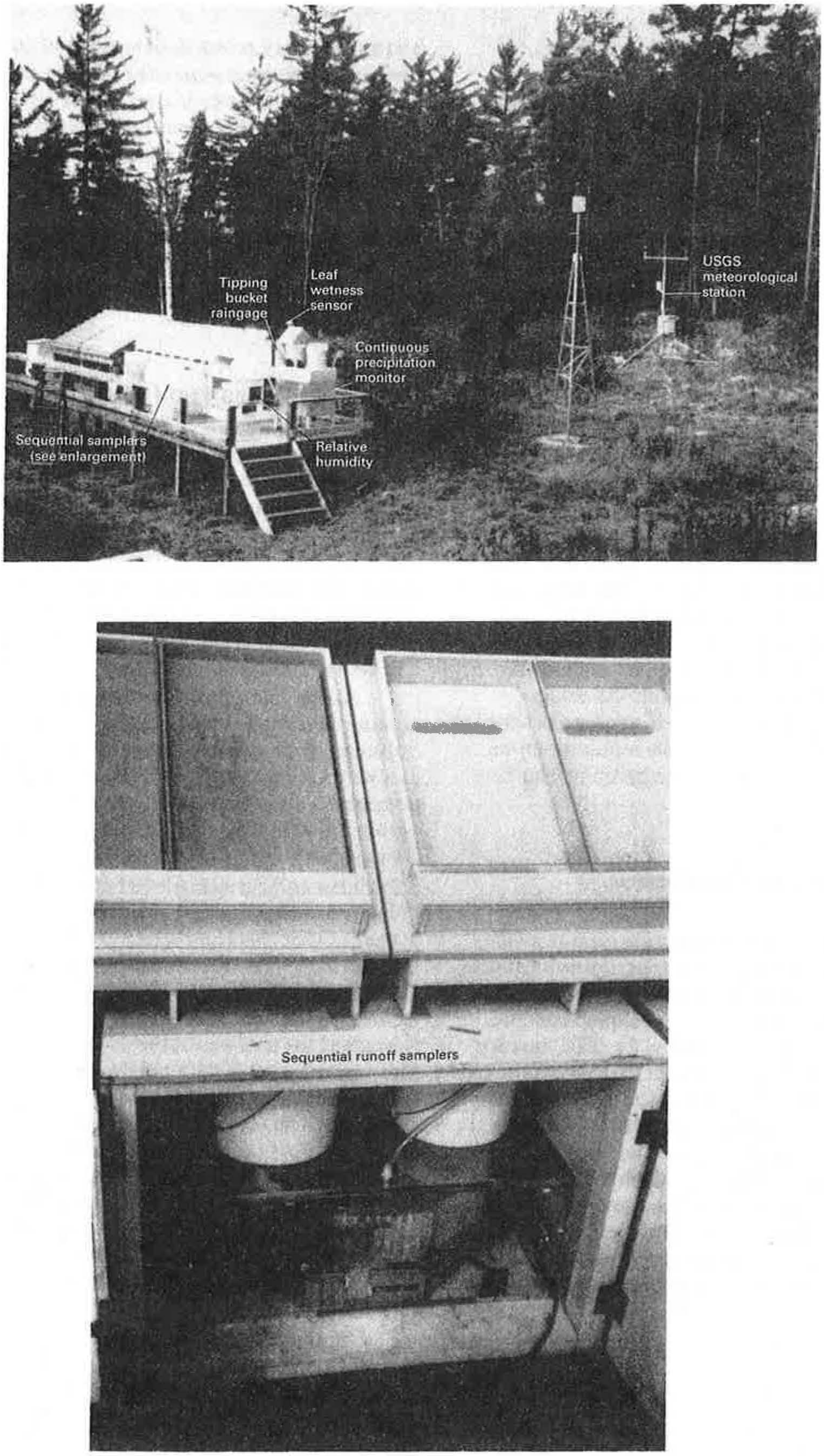

Figure 2. Instrumentation of the study site.

Meteorological Station

A Campbell Scientific meteorological station was installed at the site in 1987 (3 in fig. 2). The station was equipped with a Met-One 014A windspeed sensor, Met-One 024A wind-direction sensor, LI-200 silicon pyranometer, and a Model 201 relative humidity probe which consisted of a Phys-Chemical Research Model PCRC-11 electrohumidity sensor and a Fenwal UUT-51J1 thermistor. A CR-21 datalogger recorded data at 15-minute intervals.

Wetness Sensors

Stone dissolution has been reported to be influenced by the time of wetness (TOW) of the stone surface between storms (Ashton and Sereda, 1982). TOW measurements were made by using two types of wetness sensors and a relative humidity probe (the same model described above) (4 in fig. 2). The wetness sensors are a gypsum-coated circuit grid (5 in fig. 2, a modified Campbell model 231 leaf-wetness sensor) and a limestone-block resistor (not visible in fig. 2, adjacent to 5). A Campbell Scientific datalogger recorded data at hourly intervals. A detailed description and performance of the relative humidity probe and the two wetness sensors are given by See and others (1988).

Continuous Precipitation Monitor

The continuous precipitation monitor is a Geotech Model 0650 wet/dry precipitation collector modified for the continuous measurement and recording of precipitation amount, temperature, pH, and specific conductance during storms (6 in fig. 2). The monitor contains a rainfall-collection bucket that is covered by a lid between storms. The lid is lifted in response to water droplets accumulating on a rainfall sensor. A Campbell Scientific datalogger was programmed to record data at 1-minute intervals when the rainfall sensor became wet. Otherwise, the monitor recorded average values at 3-hour intervals and minimum and maximum values at 24-hour intervals. A detailed description and operational protocol for this monitor are given by Reddy and others (1986a).

Ancillary Measurements

NADP/NTN has been collecting weekly wet-only precipitation samples at the site since October 1978. Wet-only refers to precipitation collected from samplers that are exposed only during rainfall. NYDEC and the State University of New York record climatological and meteorological data simultaneously at the study site and also on a 30-m tower (above tree canopy) located about 0.8 km north of the HIF meteorological station at the study site (3 in fig. 2). NYDEC also recorded data from CSI 1600 nitrogen oxide, TECO 43 sulfur dioxide, and TECO 49 ozone gaseous analyzers on an hourly basis at the time of this study (Shores and others, 1987).

Sample Collection and Analysis

Seven summer storms were selected for study during July 1987 through September 1989. The selection was based on the ability of onsite personnel to anticipate the start of a storm and to ensure that all instrumentation (table 1) was activated and operational. The selection of these seven storms was considered random and was based on no criteria other than the preparedness of the field personnel. In all 238 samples were collected.

All sample preparation, analyses, and quality control were made by onsite and laboratory personnel. Subsamples of aggregated sequential samples were analyzed onsite for pH, specific conductance, and temperature as soon as possible after collection, typically within a few hours. The remainder of the aggregated samples were filtered through 0.45-µm pore-size Schleicher and Schuell filters (grade OE67) into clean, polyethylene bottles and sent to the project laboratory at the U.S. Geological Survey National Water Quality Laboratory in Arvada, Colo., for analysis of major ions. For quality control, field blanks and Standard Water Reference Samples (SRWS) were processed onsite and submitted for analysis at the same time as runoff samples. Blanks and SRWS also were processed and analyzed in the laboratory on a regular basis. Major anion concentrations (sulfate, nitrate, and chloride) were determined by ion chromatography using a Dionex Model QIC. Major cation concentrations (calcium, magnesium, and sodium) were determined by inductively coupled plasma spectrophotometry using a Jarrell-Ash Model 975. Alkalinity was determined using a Radiometer Acid Rain Analysis System (ARAS) automated titrator.

Analytical precision for SRWS and replicate samples typically were better than 10-percent relative standard deviation. SRWS were always within two standard deviations of the most probable value. In general, results for blank samples were less than the detection limits of the methods used. Analytical method detection limits are given by Fishman and Friedman (1985) and Garbarino and Taylor (1979). As an additional check on analytical accuracy, major ion balances were calculated for all samples and typically were within 10 percent for carbonate-stone runoff samples and 20 percent for glass runoff samples.

CHEMICAL CHARACTERISTICS OF RAINFALL AND RUNOFF

Rain rate and chemical concentrations from the carbonate-stone and glass runoff fluctuated three- to tenfold during storms. Net calcium-ion concentrations from the carbonate-stone runoff, a measure of stone dissolution, typically fluctuated twofold during these storms. At the start of the storms studied, concentrations of sulfate and other major ions (chloride, nitrate, calcium, magnesium, and sodium) in the glass runoff were increased and indicate either washoff of accumulated dry deposition on the glass microcatchment or washout from the atmosphere, or both. Fluctuations in the concentrations of major ions in the glass runoff seem to be related to changes in rain rate; concentrations of major ions were high at the start of episodes of high rain rate and then decreased through the high rain-rate episode, which indicates the importance of atmospheric washout processes. Results from seven selected summer storms are listed in the Appendix[1], and important characteristics are summarized in table 2.

Rain rate and runoff chemistry during Storm 5 are shown in figure 3. Net concentration values were calculated by subtracting the glass runoff concentrations from the carbonate-stone runoff concentrations. Net calcium corresponds to calcium dissolved from the stone surface by incident rainfall. Calculation of net

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[1] Also available on computer disk.

Table 2. Summary of important characteristics of seven summer storms in the Adirondack Mountains, New York, 1987–89

[m/s, meters per second; meq/L, milliequivalents per liter; mm/hr, millimeters per hour; mm, millimeters; µg/m^2, micrograms per square meter; µg, micrograms; NA, data not available]

Storm	1	2	3	4	5	6	7
Dry period prior to storm (days)	8	10	<1	6	7	2	12
Average surface wind direction[1]	SW	ESE	WSW	SE	V	V	SE
Average windspeed (m/s)	3.5	3.3	4.5	8.4	4.6	7.4	0.54
Net sulfate (meq/L)[2] (first effective runoff sample)	0.015	0.018	0.023	0.024	0.038	0.06	–0.05
Net calcium (meq/L) (first effective runoff sample)	0.43	0.54	0.32	0.03	0.34	0.22	1.06
Initial pH (first sequential sample)	4.51	4.18	4.88	4.13	4.69	3.88	3.34
Minimum pH (glass runoff)	4.51	4.18	4.49	4.09	3.98	3.88	3.34
Maximum pH (glass runoff)	4.86	5.38	4.91	5.12	4.89	4.77	4.25
Average pH (glass runoff)	4.69	4.88	4.67	4.54	4.43	4.34	3.75
Total hydrogen ion load (meq/L)	0.15	0.15	0.14	0.57	0.18	0.66	0.94
Total hydrogen ion load (meq/L)[3]	0.11	0.10	0.14	0.47	0.12	NA	NA
Maximum rainfall rate (mm/hr)	22.9	15.2	45.7	11.4	15.2	91.4	2.5
Average rainfall rate (mm/hr)	9.5	3.1	8.4	4.1	6.3	27.1	1.4
Total rain depth (mm)	7.43	11.22	7.15	24.04	6.67	20.29	4.71
Duration of storm (hours)	7	8	14	33	7	4	7
Total dissolution (µg/m^2)	0.117	0.177	0.104	0.294	0.102	0.139	0.120
Total recession (µm)[4]	0.043	0.066	0.038	0.109	0.038	0.051	0.044

[1] SW = Southwest; ESE = East-Southeast; WSW = West-Southwest; SE = Southeast; V = Variable.

[2] Effective runoff was determined by identifying the first high rain rate resulting in the first runoff effectively flushing the stone surface.

[3] Total hydrogen ion loads from the continuous precipitation monitor.

[4] Dissolution expressed in terms of stone surface vertical recession in units of micrometers (µm) per storm.

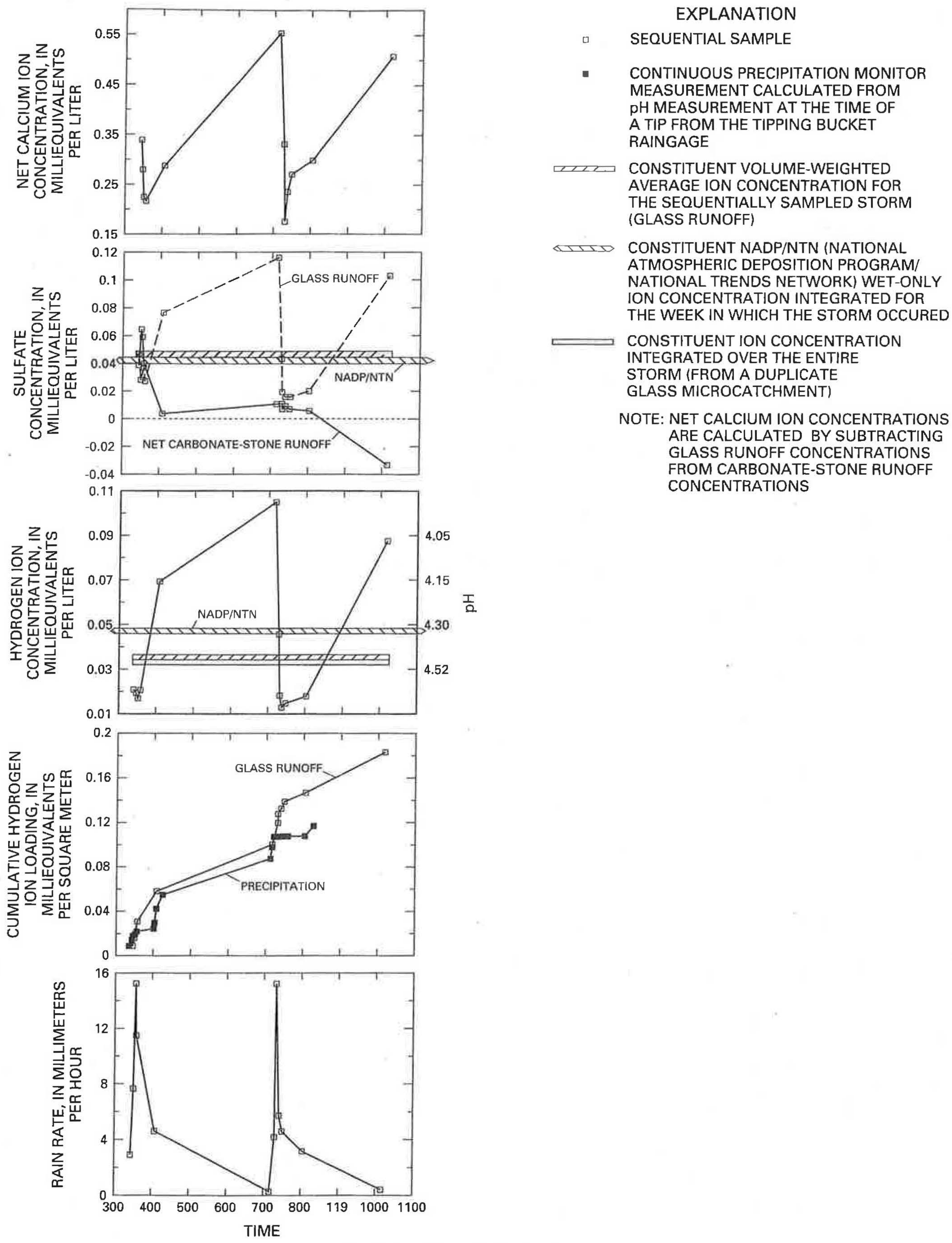

Figure 3. Rain rate and rainfall-runoff chemistry during Storm 5, September 13, 1988, Adirondack Mountains, New York.

concentrations was adopted to reflect only the changes caused by interaction of the stone surface with rainfall. Subtraction of glass runoff concentrations removed the contribution of particulate dryfall and the incident rain chemistry from the concentrations in the stone runoff. Dry deposition of gaseous pollutants will be different for the glass surface and the stone surface. Literature and laboratory studies (Baedecker and others, 1990; Reddy, 1989; Sherwood and Reddy, 1988) suggest that sulfur dioxide will accumulate on wet carbonate stone surfaces, but not on glass surfaces. This accumulation involves the formation of solid calcium sulfate compounds on the stone surface through the mediation of water.

Also shown in figure 3 is the glass-runoff constituent volume-weighted average ion concentration for the sequentially sampled storm, the constituent ion concentration integrated over the entire storm from a duplicate glass microcatchment, and the constituent NADP/NTN wet-only ion concentration integrated for the week in which the storm occurred. The volume-weighted average ion concentration for the sequential samples should equal the ion concentration of a single sample collected over the entire storm. While the NADP/NTN rain chemistry is not directly comparable to the sequentially sampled rain chemistry because it is wet-only and typically represents more than one storm, the NAPD/NTN data can serve as a reference level of precipitation chemistry over a standard 1-week interval, showing whether the rain chemistry during this period is atypical. The NADP/NTN data for the 1-week interval in which Storm 5 occurred (fig. 3) was for only one storm (Storm 5). Table 2 indicates that, with the exception of Storms 3 and 4, the duration of each storm was 4–8 hours. Surface wind direction, total rain depth, and length of dry period prior to each storm were variable.

Rain Rate and Runoff Chemistry

Rain rate fluctuated from 0.1 to 91 mm/hr during the seven storms studied. Onsite observations indicate that the nature of summer storms in the Adirondacks during the study period was such that episodes of high rain rate (cloudbursts) were frequent (C. Demers, Adirondack Ecological Center, oral commun., 1990). These cloudbursts were short in duration, typically occurring within 10 to 30 percent of the total time of the storm, and commonly were separated by longer episodes of low rain rate (fig. 3). Episodes of high rain rate resulted in an increased runoff rate from the carbonate-stone surface and episodes of low rain rate resulted in a decreased runoff rate from the carbonate-stone surface. Episodes of low rain rate (less than 5 mm/hr) were responsible for 13 to 76 percent of the total carbonate-stone runoff volume. At the start of a storm, rainfall did not run off the carbonate stone until the stone surface became sufficiently wetted. For example, at the start of Storm 2, rain rate was low, which resulted in the advancement of the sequential samplers with little or no runoff collected.

The appendix lists the runoff volumes collected from the sequential samplers. Runoff volumes were not recorded during Storms 1 and 2, necessitating the calculation of volumes from the rainfall depth and catchment area. Glass and marble runoff volumes recorded for Storms 3 through 7 generally were 15 percent higher than calculated volumes. One possibility for the differences between measured and calculated volumes is the collection efficiency of the catchment. For example, wind-driven rain will cause variations in collection volumes. Differences between glass and marble runoff volumes were generally within 20 percent.

Fluctuations in Concentrations of Hydrogen, Sulfate, and Nitrate Ions

Hydrogen-ion, determined from pH values, and sulfate-ion, and nitrate-ion concentrations followed similar trends during all seven storms. Figure 3 and table 2 indicate that rainfall (glass runoff) pH can fluctuate by more than 1 unit and generally remains less than 5.0 during a storm. For all but Storms 3 and 5, minimum rainfall pH occurred at the start of the storm, generally followed by an increase in rainfall pH as the storm progressed. Values of rainfall pH remained at increased levels except when rain rate decreased, during which time rainfall pH values generally began to decrease (see appendix). Four storms indicated that minimum rainfall pH often was coincident with the start of the storm or with episodes of low rain rate. Generally, maximum rainfall pH often was coincident with or immediately followed episodes of maximum rain rate. Similar variations among hydrogen-, sulfate-, and nitrate-ion concentrations with changing rainfall rate during a storm indicate that acid species that produce acid rain in the Adirondack Mountains of New York are sulfur and nitrogen acids derived from sulfur dioxide and nitrogen oxide pollutants.

Effects of Sampling Methods

In this section, a comparison of average ion concentration values for each storm are calculated using data from three sampling methods: (1) volume-weighted average ion concentration for the sequentially sampled storm, (2) ion concentration, integrated for the entire storm, from a duplicate glass microcatchment, and (3) NADP/NTN wet-only ion concentration integrated for the week during which the storm occurred.

In addition to the NADP/NTN samples being wet-only, each sampling method differed in sampling interval. Sequential samples were collected after every 0.254 mm of rainfall during a storm. Storm-integrated samples were collected at the end of a storm. NADP/NTN integrated samples were collected weekly. In all but Storm 5, the NADP/NTN samples represent composites of the storms sequentially sampled in this study plus at least one more storm. Hydrogen- and sulfate-ion concentrations for the storms studied typically differed by zero to fivefold among the three sampling methods (table 3).

Sequential sampling of runoff from carbonate-stone (marble) and glass (reference) microcatchments provided a detailed record of the fluctuations of rainfall and runoff chemistry during a storm and indicated episodes or pulses of acidic rainfall that may cause accelerated dissolution to carbonate-stone building materials (Schuster and Reddy, 1988, 1989). Sequential sampling discloses the coincidence between episodes of low rain rate and decreased rainfall pH, which result in acidic pulses of rainfall. These pulses of acidity, occurring during the course of a storm, are obscured in integrated storm samples or weekly NADP/NTN wet-only samples (see fig. 3) that represent average chemistry of rainfall runoff or precipitation for a specified period. In some circumstances, weekly NADP/NTN samples were more acidic than the average sequential samples or integrated samples. However, even in these cases, acid pulses within storms were still more acidic than the NADP/NTN samples. It appears that the NADP/NTN precipitation chemistry data consistently underestimate rainfall acid peaks encountered during storms at this site and possibly in other acid-rain-affected regions.

Variation among sampling methods indicates that the sampling interval is an important factor in considering the effect of storm chemistry fluctuations on the stone surface. The differences in concentrations of hydrogen and sulfate between the sequential samples and the NADP/NTN samples probably result from the integration of more than one storm in the NADP/NTN samples. Sequential samples are believed to be the most useful for interpreting dissolution of carbonate stone caused by acid rain.

Table 3. Differences in hydrogen- and sulfate-ion concentrations in samples collected by three sampling methods

[meq/L, milliequivalents per liter; NA, data not available]

Storm	Sampling method[1]	Average hydrogen-ion concentration (meq/L)	Average sulfate-ion concentration (meq/L)
1	A	0.021	0.018
1	B	NA	NA
1	C	0.09	0.10
2	A	0.012	0.006
2	B	0.022	0.021
2	C	NA	0.029
3	A	0.021	0.032
3	B	0.014	0.028
3	C	NA	0.07
4	A	0.028	0.028
4	B	0.017	0.022
4	C	0.059	0.068
5	A	0.036	0.045
5	B	0.034	0.045
5	C	0.045	0.042
6	A	0.05	0.045
6	B	0.05	0.045
6	C	0.048	0.042
7	A	0.216	0.189
7	B	NA	NA
7	C	0.0475	0.0393

[1] A = Glass runoff volume-weighted average ion concentration for the sequentially sampled storm; B = Glass runoff ion concentration from a duplicate glass microcatchment integrated for the entire storm; C = NADP/NTN wet-only ion concentration for the week during which storm occurred.

Cumulative Hydrogen-Ion Loading

Hydrogen-ion loading was calculated by multiplying the hydrogen-ion concentration, determined from rainfall pH values, by the volume of rainfall collected over the area of a microcatchment during the time of three tips from the tipping-bucket raingage. Figure 3 illustrates that most of the hydrogen-ion loading occurred during episodes of high rain rate (increased rainfall runoff from the glass and carbonate-stone surfaces). However, the hydrogen-ion concentration generally decreased during these episodes. During episodes of high rain rate (high runoff rate), there was less contact time of water with the carbonate stone surface and less opportunity for hydrogen ions to react with the stone surface. During episodes of low rain rate, runoff rate on the carbonate-stone surface decreased, allowing an extended contact time between the rainfall runoff and the stone surface.

Two methods of measuring hydrogen ion inputs were used in this study; sequential sampling using Gilson Escargot samplers and continuous flow measurements using a continuous precipitation monitor. Cumulative hydrogen-ion loading on the glass microcatchment was compared to cumulative hydrogen-ion loading in the collection bucket from the precipitation monitor for the same storm. Table 2 shows that, with the exception of Storm 3, the total hydrogen-ion loading for the continuous precipitation monitor is consistently less by an average of 28 percent.

During Storms 1 and 2, cumulative hydrogen-ion loading calculated by the methods did not agree. Calibration data for the continuous precipitation monitor indicated the pH electrode had drifted considerably during the time of Storms 1, 2, and 5 (table 4). These data are not used in the analyses of the sequential sample chemistry but are used for the comparison among the pH and specific conductance measured sequentially and that measured using the continuous precipitation monitor. For example, from September 9 to 15, the time in which Storm 5 occurred, measurement of the standards indicate the monitor had a pH drift of 0.5 to 1.0 unit. Storm 5 occurred on September 13, and pH from the sequential sampler and the monitor indicate good agreement over much of the storm, which suggests that the pH drift in the monitor occurred after September 13.

Major differences between sequential-sample rainfall pH and continuous precipitation monitor pH are associated with high rain-rate episodes. The continuous monitor is a wet-only collector, which may explain why total hydrogen-ion loading, calculated from the monitor, is 28 percent less than that calculated by the sequential-sampling method. This reasoning, however, does not explain the elevated concentrations of hydrogen and sulfate ions in the wet-only NADP/NTN collector (table 3). Integration of more than one storm over the week in which sequential samples were taken from an individual storm was most likely responsible for differences in concentrations between sampling methods. Laquer (1990b) compared continuous-flow and sequential-sampling methods for determining rainfall pH and conductivity and concluded that, although specific conductance was comparable between the two systems, flow-system pH measurements were biased toward excess acidity probably because of insufficient electrode equilibration time or streaming potentials. These factors are especially important for low ionic strength samples. Matsumoto and others (1988) and Reddy and others (1985) also

Table 4. Calibration data for pH from the continuous precipitation monitor before and after five summer storms in the Adirondack Mountains, New York, 1987–88

[NA, data not available]

Standard	Storm number									
	1		2		3		4		5	
	Calibration		Calibration		Calibration		Calibration		Calibration	
	Before	After	Before	After	Before	After	Before	After	Before	After
	7/23/87	7/29/87	9/4/87	9/11/87	7/27/88	9/6/88	7/27/88	9/6/88	9/9/88	9/15/88
[1]4 buffer	3.96	3.28	3.99	4.12	4.02	4.04	4.02	4.04	4.02	4.40
[2]7 buffer	7.03	7.20	7.01	7.14	7.04	7.03	7.04	7.03	7.00	6.98
[3]H_2SO_4	3.64	NA	4.06	4.37	4.01	4.06	4.01	4.06	4.02	4.52
[4]H_2SO_4	3.16	NA	3.05	3.26	3.05	3.05	3.05	3.05	3.03	3.57
[5]DI water	5.44	NA	5.39	7.82	6.06	6.41	6.06	6.41	5.57	6.99

[1] Phthalate pH buffer solution (pH = 4.00 at 25 degrees Celsius).

[2] Phosphate pH buffer solution (pH = 7.00 at 25 degrees Celsius).

[3] Sulfuric acid standard prepared in laboratory (pH = 4.01 at 25 degrees Celsius).

[4] Sulfuric acid standard prepared in laboratory (pH = 3.03 at 25 degrees Celsius).

[5] Deionized-distilled water (pH = 5.6 at standard temperature and pressure).

reported bias toward excess acidity for the continuous-flow systems during high-rain-rate, low-temperature rainfall. Further work is necessary in the area of continuous-flow sampling methods.

EFFECTS OF ACID DEPOSITION

In the following section, we identify factors that most strongly influence the dissolution of carbonate stone. These factors include contributions of wet deposition during storms and dry deposition prior to storms. Wet deposition in the form of acid rain is quantified relative to dissolution caused by the natural solubility of carbonate stone in rain that is in equilibrium with atmospheric carbon dioxide. Also discussed is the importance of dry deposition (gaseous SO_2) and its effects on stone dissolution. Finally, a comparison to the results of other studies is presented.

Dissolution of Carbonate Stone

Concentrations of net calcium ions in the runoff from the carbonate stone were used to determine the amount of dissolution of the carbonate stone. The net calcium concentrations and therefore the dissolution of the carbonate stone fluctuated during storms, which indicated the dissolution rate of the stone during a storm was not constant.

Net calcium ion concentrations from the carbonate-stone runoff typically fluctuated twofold during the storms studied. Generally, the net calcium concentrations in the stone runoff were small at the start of the storm, increased during episodes of low rain rate, and were elevated at the onset of episodes of high rain rates. Net calcium concentrations in the runoff decreased rapidly from the elevated concentrations at the onset of episodes of high rain rate. The runoff from episodes of high rain rate (cloudbursts), contained 21 to 73 percent of the total calcium lost by the carbonate stone during the entire storm.

It is difficult to identify those factors or combinations of factors that most strongly influence the dissolution of carbonate stone. In addition to the dissolution of the stone surface by direct rainfall, gaseous atmospheric pollutants such as sulfur dioxide and, to a lesser extent, nitrogen oxides accumulate on the stone surface during the dry period prior to a storm. Antecedent carbonate-stone moisture is important for both the generation of rainfall runoff from and dry deposition to the carbonate stone surface (See and others, 1988). At present, the working hypothesis is that sulfur dioxide reacts with the carbonate stone ($CaCO_3$) in the presence of moisture, such as dew, fog, or high relative humidity, to produce a calcium sulfate mineral ($CaSO_4.2H_2O$) on the stone surface (Baedecker and others, 1990; Reddy, 1989; Sherwood and Reddy, 1988). This surface calcium sulfate mineral has a higher solubility than carbonate stone and will dissolve preferentially in the initial runoff from the stone at the start of a storm. If this hypothesis is correct, increased net calcium and sulfate concentrations would occur in the runoff at the start of the storm. Increased net calcium and sulfate concentrations, however, typically are coincident with the first high rain rate during the storm and not during the first sequential sample of the storm, which indicates that a low rain rate at the start of the storm does not sufficiently wet the stone surface to cause dissolution of surface calcium sulfate. Sufficiently long duration low rain rate would be expected to wash calcium sulfate from the stone. Thermal properties, time of day, and relative humidity will also influence the time it takes to sufficiently wet a carbonate stone (See and others, 1988). Surface calcium sulfate does not seem to be removed until a threshold quantity of water flushes the stone surface.

Figure 4 shows the effects of rain rate (mm/hr) and rainfall pH on the quantity of dissolution of the carbonate stone and indicates that episodes of low rain rate (less than 5 mm/hr) and acidic pH (about 4.0) lead to the accumulation of carbonate dissolution products on the carbonate stone surface as indicated by the increased net calcium in the runoff during these episodes (fig. 3). Past studies indicate that episodes of high rain rate increased removal of accumulated carbonate-dissolution products from the carbonate-stone surface (Baedecker, and others, 1990; Reddy, 1989; Sherwood and Reddy, 1988). Results from

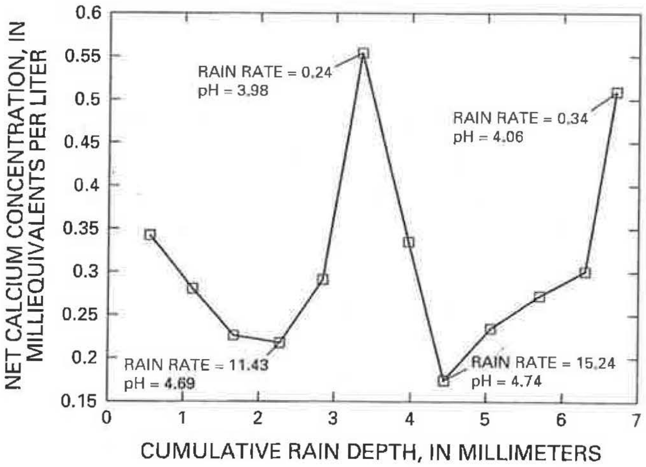

Figure 4. Concentration of net calcium ion in carbonate-stone runoff as a function of cumulative rainfall depth, Adirondack Mountains, New York, 1987–89.

sequential-runoff samples indicate that net calcium and net sulfate concentrations were consistently elevated at the onset of episodes of high rain rate but, as high rain-rate episodes progressed, accumulated carbonate dissolution products were washed from the stone surface, resulting in a rapid decrease of net calcium and net sulfate concentrations. It is more likely episodes of low rain rate and low pH that are chiefly responsible for the dissolution of carbonate stone and that high rain-rate episodes seem necessary to flush accumulated carbonate-dissolution products from the stone surface.

Although the action of rainfall seems to influence dissolution of carbonate stone during a storm, rainfall acidity is an important factor in quantifying the dissolution. Generally, rainfall pH decreased during episodes of low rain rate and at the onset of episodes of high rain rate. This increased concentration of hydrogen ion seemed to increase the dissolution of calcium carbonate, which resulted in increased calcium concentrations in the runoff during these episodes. Figure 5 separates the effects of rain rate and rainfall pH on the dissolution of the stone by normalizing the surface dissolution to the depth of rain. At any given rain rate, rainfall pH values greater than 4.0 seemed to have little or no effect on the quantity of dissolution of the stone. At rainfall pH values less than 4.0, however, the quantity of dissolution increased. This increase seemed to be greatest at rain rates less than 5 mm/hr, increasing about twofold when the rainfall pH decreased from greater than 4.5 to less than 4.0. As rain rate increased, rain pH seemed to be a less important factor in the dissolution of the stone. There is some indication that dissolution decreases slightly as rain rate exceeded values of about 5 mm/hr. High rain rates result in increased runoff rates, which allowed shorter and less efficient contact time between the rainfall and the stone surface. In addition, rainfall pH values generally increased rapidly in response to rapid increases in rain rate. Episodes of high rain rate and increased rainfall pH seem to be less effective on the dissolution of the stone than episodes of low rain rate and decreased pH. These factors have been analyzed quantitatively in another aspect of this study (Kishiyama, 1991).

Another factor influencing dissolution of the carbonate stone is the duration of the dry period prior to the storm. Longer exposure of the stone to gaseous pollutants such as sulfur dioxide during dry periods may result in a greater buildup of calcium sulfate on the surface of the stone. Net sulfate concentrations in the initial runoff, however, are low relative to the remainder of the storm in five of the seven storms examined in this study. Net calcium concentrations also are low in the initial runoff relative to the remainder of the storm in six of the seven storms studied. These low concentrations of calcium and sulfate at the start of a storm probably are because of wetting processes of the stone in which the stone must first become sufficiently wet before runoff can effectively flush calcium and sulfate from the stone surface. This causes a lag time between the start of the storm and the time that increased calcium and sulfate concentrations are observed in the runoff. The sample, collected after the stone surface had been completely wetted, was operationally defined as the "first effective runoff sample". Therefore, the first effective runoff sample should be used to assess the effects of the dry period prior to the storm. For the storm data analyzed here, the first effective runoff sample was typically that associated with the first episode of high rain rate in a given storm. This first episode of high rain rate effectively flushes the stone surface, removing atmospheric particulates, reaction residues from previous storms, and accumulated reaction products associated with episodes of low rain rate coincident with acidic rainfall.

Table 2 shows no relation between the length of the dry period prior to the storm and the quantity of net sulfate and calcium from the carbonate-stone runoff in the first effective runoff sample during the storm. One would expect accumulation of calcium sulfate by dry deposition of sulfur dioxide in the dry period prior to the storm. Uptake of sulfur dioxide by the stone should be proportional to sulfur dioxide concentration, time of wetness of the stone, and the length of the dry period prior to the storm (See and others, 1988). Length of the dry period prior to the storm and initial rainfall pH may regulate the amount of calcium and sulfate flushed from the stone surface in the first effective runoff. For example, Storm 7 had the longest dry period prior to the storm (12 days), the lowest initial glass runoff pH (3.34), and the highest net calcium concentration in the first effective runoff from the carbonate stone (1.06 meq/L). Storm 2, however, had a dry period prior to the storm of 10 days, an initial glass-runoff pH of 4.18, and a net calcium concentration in the first effective runoff from the carbonate stone of only 0.54 meq/L. A method is currently being developed to separate the contributions of sulfur dioxide dry deposition and hydrogen ion to the dissolution of the stone during the initial rainfall episode.

Comparison to Results of Other Studies

Johnsson and Reddy (1990) using the continuous precipitation monitor at the Adirondack Ecological Center in New York found that rainfall pH varied throughout the course of summer storms; low pH values at the beginning of a storm increased as rainfall rate

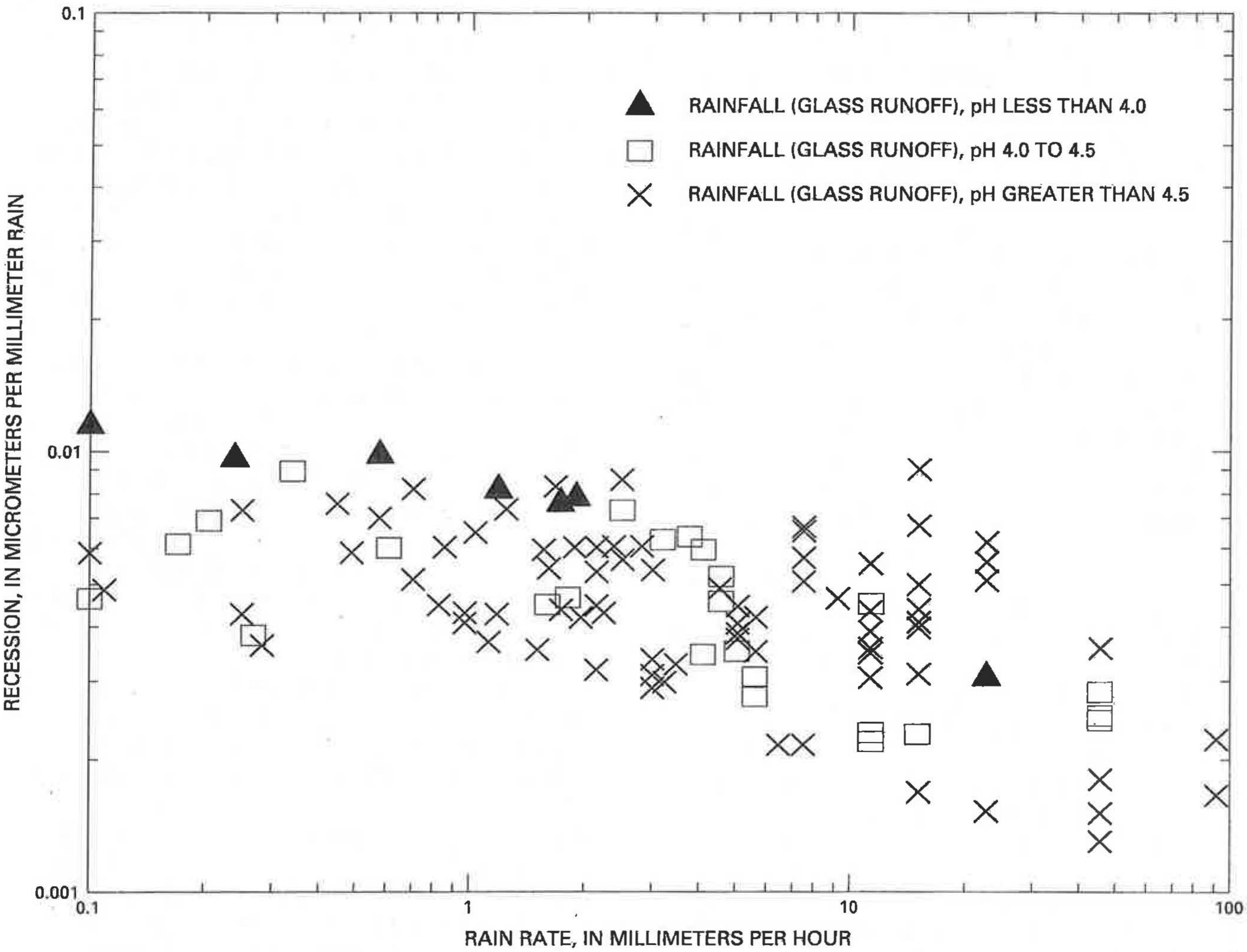

Figure 5. Rainfall pH stratified recession expressed in terms of micrometers lost from the carbonate-stone surface as a function of rain rate, Adirondack Mountains, New York, 1987–89.

increased. Borchert (1986) reported rainfall acidity increased as rainfall rate decreased during storms. Kins (1982) determined that concentrations of major ions decreased with increasing rainfall rate during convective storms.

Reddy (fig. 4, 1989) gives the predicted carbonate-stone (marble) dissolution for three rainfall pH classes of integrated storm samples from 1985–87. Figure 6 shows predicted carbonate-stone dissolution from Reddy (1989) superimposed on the cumulative dissolution of the carbonate stone as a function of cumulative rain depth on the carbonate-stone microcatchment. The predicted carbonate-stone dissolution for the pH class less than 4.0 closely compares to the

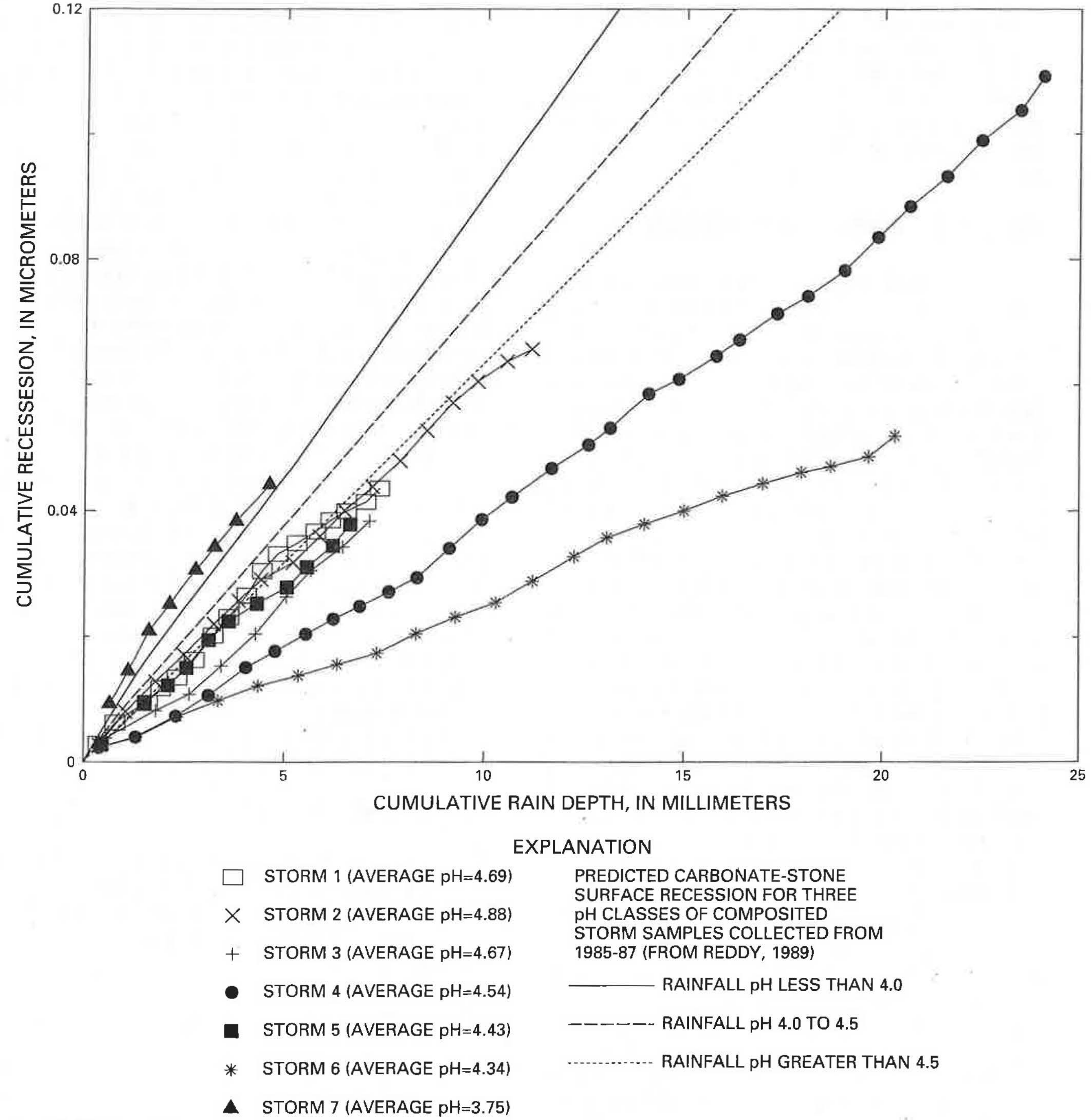

Figure 6. Cumulative recession expressed in terms of micrometers lost from the carbonate-stone surface as a function of cumulative rain depth on the carbonate stone, Adirondack Mountains, New York, 1987–89.

carbonate-stone dissolution line from Storm 7 which had an average rainfall pH of 3.75. Storms 1, 2, 3, and 5, with average rainfall pH's greater than or close to 4.5, compare to the predicted carbonate-stone dissolution for the rainfall pH class greater than 4.5. Storms 4 and 6, however, show dissolutions that are lower than the predicted carbonate-stone dissolutions for their pH classes. Dissolution for these storms was calculated by using measured and calculated runoff volumes which resulted in insignificant differences. Storm 6 had an average rainfall rate that was at least three times higher than the other storms, which could have caused a less efficient contact time of the rain with the stone surface and explain why Storm 6 had the lowest dissolution rate. Dissolution for Storms 4 and 6 are being investigated further.

SUMMARY AND CONCLUSIONS

Sequential sampling of runoff from carbonate-stone and glass microcatchments during seven selected summer storms in acid-rain-affected regions of the Adirondack Mountains in New York State provided a detailed record of the episodic fluctuations in rain rate and runoff chemistry. Rain rate and chemical concentrations from the carbonate-stone and glass runoff fluctuated three- to ten-fold during a storm. Net calcium-ion concentrations from the carbonate-stone runoff, a measure of stone dissolution, typically fluctuated twofold during these storms. At the start of a storm, concentrations of major ions in the glass runoff were increased and indicate either washoff of accumulated dry deposition on the glass microcatchment, or washout from the atmosphere, or both. Increased net sulfate and net calcium concentrations in the first effective runoff at the start of a storm indicated that atmospheric pollutants deposited on the stone surface during dry periods formed calcium sulfate minerals, an important process in carbonate stone dissolution. During storms, fluctuations in concentrations of major ions in the glass runoff seem to be related to changes in rain rate. All major ion concentrations decreased rapidly during high rain-rate episodes, which indicates the importance of washout processes during a storm.

Variations among sampling methods indicate that sampling intervals are an important factor in considering the effect of storm chemistry fluctuations on carbonate-stone surfaces. Despite these differences among weekly integrated, storm-integrated, and sequential samples, sequential samples are considered to be most useful when considering stone dissolution by acid rain. Coincident low rain rate and decreased rainfall pH result in maximum concentrations of hydrogen ions to the stone surface. These pulses of acidity are obscured in the chemistry of integrated samples but are recorded in sequential sampling during a storm.

The dissolution rate of carbonate stone typically had a twofold fluctuation during storms. Rain rate and rainfall acidity are considered to be major factors influencing the dissolution of carbonate stone. At any given rain rate, pH values generally greater than 4.0 seemed to have little effect on the quantity of dissolution of the stone. At pH values less than 4.0, however, the amount of dissolution was greatest. Dissolution accelerated, increasing about twofold, when pH of rainfall runoff decreased from greater than 4.5 to less than 4.0 during episodes when rain rate was less than 5 mm/hr. This increase was most likely in response to: (1) a decrease in pH during episodes of low rain rate, (2) more efficient rainfall reaction with the stone surface, or (3) a combination of both. As rain rate increased, pH seemed to be a less important factor in the dissolution of carbonate stone. There is some indication, however, that dissolution decreased slightly as rain rate exceeded about 5 mm/hr. This decrease was most likely in response to: (1) a rapid increase in pH during episodes of high rain rate, (2) the short contact time of rainfall with the stone surface, or (3) a combination of both. These factors have been analyzed quantitatively in another aspect of this study (Kishiyama, 1991).

Analyses of data collected in this study indicate that the most important factors causing increased dissolution of carbonate-stone building materials are coincident episodes of low rain rate and decreased rainfall pH. An increase in hydrogen-ion concentration in the precipitation reacts with the calcium carbonate in the stone, which results in an increase in calcium concentrations in the runoff. High runoff rates resulting from cloudbursts remove calcium sulfate minerals formed during dry periods prior to storms and also remove dissolution products formed in large measure by chemical weathering as a result of episodes of low rain rate and decreased rainfall pH during a storm.

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APPENDIX

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89

[mL, milliliters; µS/cm, microsiemens per centimeter at 25 degrees Celsius; meq/L, milliequivalent per liter; meq/m^2, milliequivalent per square meter; µm, micrometer; mm/hr, millimeter per hour; NA, data not available]

Storm[1]	Sequential sample number	Time of sample collection	Glass runoff, volume (mL)	Glass runoff, field pH	Glass runoff, laboratory pH	Glass runoff, field conductivity (µS/cm)	Glass runoff, alkalinity (meq/L)	Glass runoff, chloride (meq/L)	Glass runoff, nitrate (meq/L)	Glass runoff, sulfate (meq/L)
1	1-3	1447	76.65	4.51	NA	15.3	0	0.00638	0.01724	0.02713
1	4-6	1628	76.65	4.71	NA	16.3	0	0.00440	0.02449	0.03885
1	7-9	1631	76.65	4.85	NA	10.5	0	0.00273	0.01122	0.02145
1	10-12	1635	76.65	4.66	NA	11.0	0	0.00431	0.00554	0.01500
1	13-15	1640	76.65	4.67	NA	10.0	0	0.00168	0.00492	0.01538
1	16-18	1706	76.65	4.80	NA	7.9	0	0.00200	0.00057	0.00972
1	19-21	1759	76.65	4.85	NA	7.2	0	0.00300	0.00600	0.01058
1	22-24	1817	76.65	4.78	NA	9.4	0	0.00300	0.01197	0.01036
1	25-27	1936	76.65	4.86	NA	6.7	0	0.00200	0.00400	0.00313
1	28-30	2040	76.65	4.65	NA	13.3	0	0.00493	0.02328	0.01131
1	31-33	2043	76.65	4.81	NA	10.3	0	0.00378	0.03171	0.01567
1	34-36	2045	76.65	4.60	NA	14.9	0	0.00348	0.04273	0.02288
1	37-39	2048	76.65	4.56	NA	15.0	0	0.00608	0.03132	0.01882
1	40-42	2051	76.65	4.59	NA	13.1	0	0.00200	0.02554	0.01633
1	43-45	2054	76.65	4.59	NA	12.1	0	0.00297	0.01889	0.01596
1	46-48	2058	76.65	4.63	NA	10.9	0	0.00420	0.01360	0.01504
1	49-51	2138	76.65	4.74	NA	8.0	0	0.00251	0.00737	0.00939
1	52-54	2140	76.65	4.71	NA	8.6	0	0.00340	0.00668	0.00955
2	1-3	1334	76.65	4.18	NA	32.5	NA	NA	NA	NA
2	4-6	1423	76.65	4.38	NA	17.9	NA	NA	NA	NA
2	7-9	1637	76.65	4.46	NA	13.7	-0.04660	0.00494	0.01744	0.02686
2	10-12	1806	76.65	4.61	NA	10.4	-0.02280	0.01094	0.00845	0.00839
2	13-15	1850	76.65	4.56	NA	10.4	-0.02530	0.01012	0.00926	0.00857
2	16-18	1911	76.65	4.61	NA	10.0	-0.02710	0.00600	0.00987	0.00900
2	19-21	1917	76.65	4.69	NA	8.2	-0.01840	0.01493	0.00876	0.00759
2	22-24	1936	76.65	4.93	NA	5.6	-0.01070	0.00501	0.00400	0.00408
2	25-27	1959	76.65	5.03	NA	4.7	-0.00850	0.00204	0.00400	0.00280
2	28-30	2019	76.65	4.95	NA	4.9	-0.00940	0.00238	0.00400	0.00340
2	31-33	2152	76.65	5.10	NA	3.0	-0.00730	0.00215	0.00400	0.00200
2	34-36	2220	76.65	5.38	NA	2.9	-0.00690	0.00200	0.00400	0.00177
2	37-39	2244	76.65	5.07	NA	4.8	-0.00650	0.00312	0.00461	0.00501
2	40-42	2308	76.65	5.21	NA	3.7	-0.00610	0.00200	0.00400	0.00161
2	43-45	2311	76.65	5.15	NA	3.8	-0.00500	0.00200	0.00400	0.00232
2	46-48	2317	76.65	5.20	NA	3.2	-0.00510	0.00395	0.00400	0.00208
2	49-51	2320	76.65	5.28	NA	2.6	-0.00490	0.00200	0.00400	0.00137
2	52-54	2341	76.65	5.41	NA	2.5	-0.00270	0.00411	0.00400	0.00131
2	55-57	2355	76.65	5.06	NA	4.6	-0.00420	0.00300	0.00400	0.00357

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Glass runoff, calcium (meq/L)	Glass runoff, magnesium (meq/L)	Glass runoff, sodium (meq/L)	Stone runoff, volume (mL)	Stone runoff, field pH	Stone runoff, laboratory pH	Stone runoff, field conductivity (µS/cm)
1	1-3	0.03973	0.00517	0.00526	76.65	7.17	NA	37.8
1	4-6	0.06328	0.00476	0.00331	76.65	7.60	NA	50.4
1	7-9	0.04114	0.00418	0.00270	76.65	7.44	NA	31.2
1	10-12	0.01784	0.00200	0.00339	76.65	7.22	NA	21.8
1	13-15	0.01552	0.00149	0.00278	76.65	7.30	NA	25.7
	16-18	0.01064	0.00100	0.00300	76.65	7.24	NA	23.4
1	19-21	0.01516	0.00200	0.00326	76.65	7.42	NA	32.9
1	22-24	0.02544	0.00200	0.00210	76.65	7.57	NA	44.1
1	25-27	0.01043	0.01100	0.00117	76.65	7.48	NA	33.6
1	28-30	0.03182	0.00400	0.00343	76.65	7.56	NA	43.3
1	31-33	0.04135	0.00500	0.00300	76.65	7.60	NA	47.2
1	34-36	0.04270	0.00400	0.00200	76.65	7.40	NA	34.8
1	37-39	0.03044	0.00400	0.03176	76.65	7.24	NA	24.7
1	40-42	0.02303	0.00300	0.00250	76.65	7.25	NA	24.5
1	43-45	0.01495	0.00100	0.00222	76.65	7.20	NA	21.9
1	46-48	0.01083	0.00200	0.00278	76.65	7.16	NA	18.3
1	49-51	0.00749	0.00100	0.00204	76.65	7.14	NA	19.0
1	52-54	0.00763	0.00100	0.00309	76.65	7.31	NA	23.7
2	1-3	NA	NA	NA	NA	NA	NA	NA
2	4-6	NA	NA	NA	NA	7.47	NA	55.6
2	7-9	0.00600	0.00100	0.00535	76.65	7.43	NA	51.0
2	10-12	0.00600	0.00170	0.01375	76.65	7.45	NA	48.1
2	13-15	0.00400	0.00110	0.01218	76.65	7.50	NA	52.1
2	16-18	0.00300	0.00080	0.00783	76.65	7.54	NA	49.7
2	19-21	0.00800	0.00220	0.01436	76.65	7.60	NA	60.4
2	22-24	0.00300	0.00070	0.00844	76.65	7.57	NA	47.6
2	25-27	0.00300	0.00060	0.00522	76.65	7.50	NA	35.1
2	28-30	0.00250	0.00056	0.00579	76.65	7.51	NA	35.9
2	31-33	0.00250	0.00042	0.00439	76.65	7.58	NA	47.5
2	34-36	0.00250	0.00028	0.00370	76.65	7.45	NA	45.4
2	37-39	0.00300	0.00135	0.01248	76.65	7.79	NA	47.7
2	40-42	0.00250	0.00035	0.00600	76.65	7.66	NA	46.0
2	43-45	0.00250	0.00033	0.00609	76.65	7.69	NA	51.3
2	46-48	0.00250	0.00027	0.00348	76.65	7.71	NA	53.0

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Stone runoff, alkalinity (meq/L)	Stone runoff, chloride (meq/L)	Stone runoff, nitrate (meq/L)	Stone runoff, sulfate (meq/L)	Stone runoff, calcium (meq/L)	Stone runoff, magnesium (meq/L)	Stone runoff, sodium (meq/L)
1	1-3	0.35200	0.00875	0.01914	0.04117	0.37246	0.01691	0.00452
1	4-6	0.44400	0.00430	0.02632	0.05420	0.49268	0.01853	0.00300
1	7-9	0.31600	0.00299	0.01478	0.02834	0.31979	0.01215	0.00235
1	10-12	0.23600	0.00503	0.00752	0.01954	0.21676	0.00730	0.00283
1	13-15	0.29200	0.00262	0.00695	0.01943	0.27509	0.00804	0.00204
1	16-18	0.26800	0.00200	0.00539	0.01874	0.25460	0.00914	0.00222
1	19-21	0.36200	0.00200	0.00820	0.01574	0.35422	0.01035	0.00300
1	22-24	0.50800	0.00200	0.01565	0.01705	0.51617	0.01271	0.00239
1	22-27	0.42000	0.00200	0.00668	0.00810	0.40992	0.00983	0.00244
1	28-30	0.47200	0.01258	0.02798	0.03275	0.49383	0.01647	0.00861
1	31-33	0.51800	0.01173	0.04247	0.04084	0.55600	0.01772	0.00674
1	34-36	0.34600	0.00730	0.04864	0.03606	0.39075	0.01396	0.00405
1	37-39	0.24400	0.01153	0.03530	0.02317	0.25903	0.00802	0.00405
1	40-42	0.26000	0.00594	0.03276	0.02316	0.26728	0.00878	0.00361
1	43-45	0.49000	0.00678	0.02656	0.02768	0.23612	0.00968	0.00531
1	46-48	0.20400	0.00504	0.01616	0.01763	0.20622	0.00679	0.00361
1	49-51	0.22400	0.00392	0.01193	0.01669	0.21372	0.00692	0.00287
1	52-54	0.29600	0.00857	0.01371	0.02144	0.29181	0.00924	0.00322
2	1-3	NA	NA	NA	NA	NA	NA	NA
2	4-6	NA	NA	NA	NA	NA	NA	NA
2	7-9	0.48660	0.01357	0.02523	0.03564	0.54571	0.01720	0.02053
2	10-12	0.48650	0.00704	0.01506	0.02648	0.54722	0.01682	0.00474
2	13-15	0.54610	0.00888	0.01165	0.01573	0.59453	0.02131	0.00552
2	16-18	0.48690	0.01192	0.02756	0.03743	0.54668	0.04122	0.01218
2	19-21	0.56170	0.01318	0.02235	0.02805	0.61592	0.03012	0.01175
2	22-24	0.50990	0.00975	0.01302	0.01864	0.55322	0.02303	0.00835
2	25-27	0.35980	0.00608	0.00867	0.01229	0.38002	0.01655	0.01505
2	28-30	0.37660	0.00392	0.00401	0.01061	0.38769	0.01387	0.00674
2	31-33	0.05780	0.00341	0.00287	0.01276	0.53154	0.01646	0.00744
2	34-36	0.47820	0.00976	0.00394	0.02481	0.48870	0.01696	0.01644
2	37-39	0.50960	0.00390	0.00361	0.01581	0.55260	0.01710	0.00879
2	40-42	0.51370	0.00344	0.00869	0.02119	0.54439	0.02055	0.00670
2	43-45	0.59520	0.01220	0.00821	0.02309	0.61349	0.02332	0.01727
2	46-48	0.58280	0.00303	0.00591	0.01684	0.59648	0.01987	0.00774
2	49-51	0.44130	0.00188	0.00372	0.01259	0.45754	0.01600	0.00444
2	52-54	0.37750	0.01331	0.00386	0.01891	0.40205	0.01609	0.01836
2	55-57	0.26350	0.00403	0.00100	0.01346	0.26897	0.01120	0.00853

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Glass runoff, hydrogen ion (meq/L)	Glass runoff, hydrogen ion load (meq/m^2)	Cumulative hydrogen ion loading (meq/m^2)	Net calcium[2] (meq/L)	Recession (μm)	Rain depth (mm)	Micrometer recession per millimeter rain	Rain rate (mm/hr)
1	1-3	0.0309	0.01267	0.01267	0.33273	0.00255	0.41254	0.00617	0.0000
1	4-6	0.0195	0.00799	0.02066	0.42940	0.00329	0.41254	0.00797	0.4527
1	7-9	0.0141	0.00579	0.02646	0.27865	0.00213	0.41254	0.00517	15.2400
1	10-12	0.0219	0.00897	0.03543	0.19892	0.00152	0.41254	0.00369	11.4300
1	13-15	0.0214	0.00877	0.04419	0.25957	0.00199	0.41254	0.00482	9.1440
1	16-18	0.0158	0.00650	0.05069	0.24396	0.00187	0.41254	0.00453	1.7585
1	19-21	0.0141	0.00579	0.05648	0.33906	0.00260	0.41254	0.00629	0.8626
1	22-24	0.0166	0.00680	0.06329	0.49073	0.00376	0.41254	0.00911	2.5400
1	25-27	0.0138	0.00566	0.06894	0.39949	0.00306	0.41254	0.00741	0.5787
1	28-30	0.0224	0.00918	0.07812	0.46201	0.00354	0.41254	0.00857	0.7144
1	31-33	0.0155	0.00635	0.08447	0.51465	0.00394	0.41254	0.00955	15.2400
1	34-36	0.0251	0.01030	0.09477	0.34805	0.00266	0.41254	0.00646	22.8600
1	37-39	0.0275	0.01129	0.10606	0.22859	0.00175	0.41254	0.00424	15.2400
1	40-42	0.0257	0.01054	0.11660	0.24425	0.00187	0.41254	0.00453	15.2400
1	43-45	0.0257	0.01054	0.12714	0.22117	0.00169	0.41254	0.00410	15.2400
1	46-48	0.0234	0.00961	0.13675	0.19539	0.00150	0.41254	0.00363	11.4300
1	49-51	0.0182	0.00746	0.14421	0.20623	0.00158	0.41254	0.00383	1.1430
1	52-54	0.0195	0.00799	0.15221	0.28418	0.00218	0.41254	0.00527	22.8600
2	1-3	NA	NA	NA	NA	NA	NA	NA	NA
2	4-6	NA	NA	NA	NA	NA	NA	NA	NA
2	7-9	0.0347	0.02275	0.02275	0.53971	0.00413	0.66021	0.00626	0.6178
2	10-12	0.0245	0.01610	0.03885	0.54122	0.00414	0.66021	0.00628	1.5766
2	13-15	0.0275	0.01807	0.05692	0.59053	0.00452	0.66021	0.00685	1.0391
2	16-18	0.0245	0.01610	0.07302	0.54368	0.00416	0.66021	0.00630	2.1771
2	19-21	0.0204	0.01339	0.08641	0.60792	0.00465	0.66021	0.00705	7.6200
2	22-24	0.0117	0.00771	0.09412	0.55022	0.00421	0.66021	0.00638	2.4063
2	25-27	0.0093	0.00612	0.10024	0.37702	0.00289	0.66021	0.00437	1.9878
2	28-30	0.0112	0.00736	0.10760	0.38519	0.00295	0.66021	0.00447	2.2860
2	31-33	0.0079	0.00521	0.11281	0.52904	0.00405	0.66021	0.00613	0.4916
2	34-36	0.0042	0.00273	0.11555	0.48620	0.00372	0.66021	0.00564	1.6329
2	37-39	0.0085	0.00558	0.12113	0.54960	0.00421	0.66021	0.00637	1.9050
2	40-42	0.0062	0.00404	0.12518	0.54189	0.00415	0.66021	0.00628	1.9050
2	43-45	0.0071	0.00464	0.12982	0.61099	0.00468	0.66021	0.00708	15.2400
2	46-48	0.0063	0.00414	0.13396	0.59398	0.00455	0.66021	0.00689	7.6200

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Time of sample collection	Glass runoff, volume (mL)	Glass runoff, field pH	Glass runoff, laboratory pH	Glass runoff, field conductivity (µS/cm)	Glass runoff, alkalinity (meq/L)	Glass runoff, chloride (meq/L)	Glass runoff, nitrate (meq/L)	Glass runoff, sulfate (meq/L)
3	1-3	1756	72	4.88	5.24	10.6	-0.00120	0.00731	0.01902	0.02800
3	7-9	2034	76	4.51	4.57	12.0	-0.02930	0.00095	0.00231	0.02486
3	10-12	2035	89	4.84	4.87	5.6	-0.01150	0.00180	0.00150	0.01071
3	13-15	2056	83	4.91	4.96	5.0	-0.00900	0.00475	0.00150	0.01054
3	16-18	2058	84	4.68	4.76	8.3	-0.01950	0.01193	0.00016	0.02543
3	19-21	2104	83	4.64	4.71	9.8	-0.02370	0.00574	0.00120	0.02802
3	22-24	2140	NA	4.72	NA	6.9	NA	NA	NA	NA
3	25-27	2441	56	4.69	4.77	8.4	-0.01850	0.00513	0.00150	0.02746
3	28-30	739	84	4.58	4.63	10.0	-0.02900	0.00976	0.00150	0.12955
3	31-34	753	94	4.49	4.54	13.3	-0.03610	0.00764	0.00150	0.04392
4	1-3	5	98	4.13	4.19	31.9	-0.08150	0.01049	0.08422	0.06481
4	4-6	14	107	4.44	4.52	13.9	-0.03770	0.00466	0.02733	0.02761
4	7-9	23	114	4.54	4.62	10.8	-0.02830	0.00395	0.01689	0.02216
4	10-12	32	109	4.66	4.75	8.1	-0.02060	0.00347	0.01165	0.01748
4	13-15	41	114	4.75	4.86	6.7	-0.01580	0.00264	0.00793	0.01401
4	16-18	50	103	4.94	5.02	4.5	-0.01040	0.00890	0.00388	0.01072
4	19-21	58	100	5.05	5.12	3.7	-0.00790	0.00281	0.00261	0.01018
4	22-24	111	99	5.03	5.16	4.2	-0.00700	0.00304	0.00334	0.01203
4	25-27	126	95	5.09	5.16	3.6	-0.00690	0.00251	0.00209	0.01067
4	28-30	141	97	5.10	5.15	3.3	-0.00720	0.00538	0.00134	0.01034
4	31-33	156	99	5.12	5.16	5.9	-0.00670	0.00180	0.00045	0.00942
4	34-36	217	108	4.84	4.97	5.0	-0.01320	0.00180	0.00435	0.01286
4	37-39	235	93	4.75	4.84	6.9	-0.01820	0.00535	0.00741	0.01763
4	40-42	256	95	4.63	4.68	9.4	-0.02440	0.00533	0.00912	0.02467
4	43-45	350	110	4.61	4.66	9.0	-0.02470	0.00555	0.00838	0.02435
4	46-48	443	109	4.73	4.82	6.6	-0.01740	0.00180	0.00489	0.01714
4	49-51	447	106	4.71	4.77	7.1	-0.02030	0.00580	0.00569	0.01889
4	52-54	451	119	4.66	4.71	8.3	-0.02270	0.00180	0.00696	0.02176
4	55-57	455	91	4.54	4.56	10.7	-0.03180	0.00180	0.01079	0.01860
4	58-60	459	107	4.74	4.78	6.8	-0.01890	0.00265	0.00544	0.01834
4	61-63	537	107	4.82	4.86	7.0	-0.01560	0.00180	0.00325	0.01507
4	64-66	624	111	4.83	4.90	5.7	-0.01340	0.00180	0.00283	0.01636
4	67-69	654	111	4.69	4.73	7.7	-0.02050	0.00233	0.00420	0.02118
4	70-72	722	85	4.46	4.52	12.1	-0.03400	0.00376	0.00841	0.03906
4	73-75	1141	114	4.26	4.28	18.5	-0.05920	0.00351	0.02301	0.05032
4	76-78	10	81	4.09	4.10	27.8	-0.09170	0.00377	0.04437	0.06795
4	79-81	744	62	4.16	NA	24.9	NA	0.00407	0.04370	0.06618
4	82-84	756	97	4.19	4.23	22.2	-0.06710	0.00362	0.03749	0.05415
4	85-87	821	89	4.22	4.26	20.7	-0.06190	0.00306	0.02540	0.05180
4	88-90	848	64	4.53	4.45	13.6	-0.03960	0.00337	0.01506	0.03317

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Glass runoff, calcium (meq/L)	Glass runoff, magnesium (meq/L)	Glass runoff, sodium (meq/L)	Stone runoff, volume (mL)	Stone runoff, field pH	Stone runoff, laboratory pH	Stone runoff, field conductivity (μS/cm)
3	1-3	0.01500	0.00290	0.0169	68	7.29	7.16	30.4
3	7-9	0.00940	0.00150	0.0063	73	7.43	7.10	23.7
3	10-12	0.00330	0.00050	0.0056	89	6.39	7.12	25.1
3	13-15	0.00230	0.00050	0.0052	92	7.31	7.12	22.1
3	16-18	0.00430	0.00070	0.0123	90	7.45	7.37	39.4
3	19-21	0.00470	0.00070	0.0063	97	7.62	7.42	41.0
3	22-24	NA	NA	NA	88	7.75	7.63	48.7
3	25-27	0.00540	0.00080	0.0086	68	7.76	7.63	47.8
3	28-30	0.00270	0.00050	0.0037	91	7.69	7.39	30.1
3	31-34	0.00300	0.00050	0.0033	74	7.70	7.54	42.2
4	1-3	0.03020	0.00730	0.01230	72	7.05	7.18	33.9
4	4-6	0.00630	0.00140	0.00023	99	7.18	7.18	28.7
4	7-9	0.00570	0.00110	0.00023	106	7.20	7.17	26.3
4	10-12	0.00460	0.00070	0.00023	95	7.30	7.04	28.1
4	13-15	0.00300	0.00050	0.00023	110	7.33	7.09	30.2
4	16-18	0.00200	0.00030	0.00023	84	7.38	7.06	25.3
4	19-21	0.00310	0.00040	0.00023	86	7.35	7.07	24.6
4	22-24	0.00500	0.00070	0.00023	81	7.38	7.03	22.7
4	25-27	0.00380	0.00040	0.00023	75	7.27	7.00	21.3
4	28-30	0.00370	0.00040	0.00023	83	7.34	7.01	19.7
4	31-33	0.00290	0.00030	0.00023	82	7.36	7.08	22.7
4	34-36	0.00370	0.00040	0.00023	96	7.46	7.29	37.3
4	37-39	0.00410	0.00040	0.00023	91	7.28	7.35	38.1
4	40-42	0.00340	0.00040	0.00023	90	7.52	7.22	29.9
4	43-45	0.00270	0.00030	0.00023	110	7.48	7.09	31.2
4	46-48	0.00140	0.00010	0.00023	105	7.53	7.09	27.9
4	49-51	0.00150	0.00020	0.00023	67	7.51	7.17	29.3
4	52-54	0.00240	0.00030	0.00023	109	6.62	7.29	37.4
4	55-57	0.00320	0.00040	0.00023	87	7.46	6.98	21.0
4	58-60	0.00160	0.00010	0.00023	105	7.48	7.10	27.5
4	61-63	0.00110	0.00010	0.00023	67	7.50	7.20	28.4
4	64-66	0.00140	0.00010	0.00023	109	7.54	7.20	28.4
4	67-69	0.00220	0.00020	0.00023	87	7.49	7.13	22.8
4	70-72	0.00510	0.00050	0.00023	105	7.47	7.27	30.2
4	73-75	0.00650	0.00060	0.00023	99	7.54	7.47	40.7
4	76-78	0.00800	0.00090	0.00023	93	7.58	7.36	39.8

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Stone runoff, alkalinity (meq/L)	Stone runoff, chloride (meq/L)	Stone runoff, nitrate (meq/L)	Stone runoff, sulfate (meq/L)	Stone runoff, calcium (meq/L)	Stone runoff, magnesium (meq/L)	Stone runoff, sodium (meq/L)
3	1-3	0.38460	0.00661	0.01054	0.03312	0.39710	0.01910	0.0106
3	7-9	0.29220	0.00820	0.00649	0.03794	0.33210	0.01280	0.0061
3	10-12	0.29490	0.00644	0.01735	0.03407	0.32170	0.02970	0.0052
3	13-15	0.27800	0.00560	0.00305	0.02570	0.28840	0.01880	0.0029
3	16-18	0.46260	0.00658	0.02969	0.05992	0.50460	0.03970	0.0040
3	19-21	0.46680	0.00720	0.03147	0.07194	0.52080	0.05550	0.0030
3	22-24	0.64150	0.00587	0.01950	0.04412	0.66590	0.04010	0.0031
3	25-27	0.63220	0.00493	0.01570	0.04570	0.66200	0.03130	0.0038
3	28-30	0.40400	0.00367	0.00127	0.04329	0.43940	0.01590	0.0026
3	31-34	0.51930	0.01499	0.02078	0.07235	0.57360	0.03860	0.0105
4	1-3	0.31460	0.00617	0.03015	0.06471	0.43100	0.01460	0.0123
4	4-6	0.29960	0.00625	0.02016	0.05207	0.03160	0.01480	0.0037
4	7-9	0.28960	0.00673	0.01408	0.03897	0.34830	0.01480	0.0035
4	10-12	0.33520	0.00595	0.01232	0.03665	0.36790	0.01330	0.0033
4	13-15	0.37320	0.00470	0.01114	0.03311	0.40310	0.01410	0.0027
4	16-18	0.30980	0.00625	0.01193	0.03121	0.33520	0.01580	0.0023
4	19-21	0.29860	0.00625	0.01193	0.03121	0.31800	0.01600	0.0029
4	22-24	0.28240	0.00495	0.00868	0.02742	0.30070	0.01220	0.0023
4	25-27	0.26360	0.00407	0.00656	0.02694	0.28280	0.01050	0.0023
4	28-30	0.24910	0.00344	0.00284	0.02357	0.26400	0.00870	0.0017
4	31-33	0.28740	0.00424	0.00876	0.02854	0.30410	0.01560	0.0029
4	34-36	0.43970	0.00554	0.02201	0.05730	0.48290	0.03530	0.0035
4	37-39	0.47940	0.00442	0.01642	0.04617	0.51300	0.02350	0.0031
4	40-42	0.36700	0.00396	0.01477	0.04277	0.40360	0.01590	0.0029
4	43-45	0.37190	0.00401	0.01559	0.04857	0.40520	0.01870	0.0023
4	46-48	0.34890	0.00302	0.00993	0.03210	0.36810	0.01310	0.0014
4	49-51	0.37330	0.00327	0.01003	0.03194	0.38640	0.01330	0.0018
4	52-54	0.48320	0.00334	0.01161	0.03760	0.50150	0.01810	0.0017
4	55-57	0.24760	0.00305	0.00727	0.04189	0.27690	0.00970	0.0037
4	58-60	0.32640	0.00348	0.01263	0.04003	0.34970	0.01950	0.0023
4	61-63	0.37190	0.00331	0.00733	0.02982	0.38230	0.01600	0.0023
4	64-66	0.38160	0.00505	0.00150	0.03007	0.38650	0.01400	0.0032
4	67-69	0.03760	0.00271	0.00304	0.02307	0.31780	0.01040	0.0020
4	70-72	0.39490	0.00248	0.00515	0.02829	0.40750	0.01320	0.00017
4	73-75	0.54350	0.00295	0.00680	0.03403	0.55690	0.01680	0.0021
4	76-78	0.52810	0.00334	0.01006	0.04163	0.54140	0.01670	0.0018
4	79-81	0.40520	0.00284	0.00823	0.03957	0.42760	0.01360	0.0016
4	82-84	0.56140	0.00312	0.00929	0.04935	0.59480	0.01630	0.0022
4	85-87	0.40070	0.00309	0.00150	0.04912	0.42650	0.02180	0.0028
4	88-90	0.72520	0.00456	0.01402	0.06644	0.76650	0.02230	0.0030

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Glass runoff, hydrogen ion (meq/L)	Glass runoff, hydrogen ion load (meq/m^2)	Cumulative hydrogen ion loading (meq/m^2)	Net calcium (meq/L)	Recession (µm)	Rain depth (mm)	Micrometer recession per millimeter rain	Rain rate (mm/hr)
3	1-3	0.0132	0.00865	0.00865	0.38210	0.00259	0.58570	0.00443	0.2569
3	7-9	0.0309	0.02027	0.02892	0.32270	0.00235	0.62877	0.00374	0.2894
3	10-12	0.0145	0.00948	0.03840	0.31840	0.00283	0.76658	0.00369	45.7200
3	13-15	0.0123	0.00807	0.04647	0.28610	0.00263	0.79242	0.00332	2.1771
3	16-18	0.0209	0.01371	0.06018	0.50030	0.00450	0.77519	0.00580	22.8600
3	19-21	0.0229	0.01503	0.07521	0.51610	0.00500	0.83549	0.00598	7.6200
3	22-24	0.0191	0.01250	0.08771	0.66590	0.00585	0.75797	0.00772	1.2700
3	25-27	0.0204	0.01339	0.10110	0.65660	0.00446	0.58570	0.00761	0.2526
3	28-30	0.0263	0.01725	0.11835	0.43670	0.00397	0.78381	0.00506	0.1094
3	31-34	0.0324	0.02123	0.13958	0.57060	0.00422	0.63738	0.00662	3.2657
4	1-3	0.0741	0.04863	0.04863	0.40080	0.00288	0.62016	0.00465	11.4300
4	4-6	0.0363	0.02382	0.07245	0.02530	0.00025	0.85271	0.00029	5.0800
4	7-9	0.0288	0.01892	0.09137	0.34260	0.00363	0.91301	0.00397	5.0800
4	10-12	0.0219	0.01435	0.10572	0.36330	0.00345	0.81826	0.00421	5.0800
4	13-15	0.0178	0.01167	0.11739	0.40010	0.00440	0.94746	0.00464	5.0800
4	16-18	0.0115	0.00753	0.12492	0.33320	0.00280	0.72351	0.00386	5.0800
4	19-21	0.0089	0.00585	0.13076	0.31490	0.00270	0.74074	0.00365	5.7150
4	22-24	0.0093	0.00612	0.13688	0.29570	0.00239	0.69767	0.00343	3.5169
4	25-27	0.0081	0.00533	0.14222	0.27900	0.00209	0.64599	0.00323	3.0480
4	28-30	0.0079	0.00521	0.14743	0.26030	0.00216	0.71490	0.00302	3.0480
4	31-33	0.0076	0.00498	0.15240	0.30120	0.00247	0.70629	0.00349	3.0480
4	34-36	0.0145	0.00948	0.16189	0.47920	0.00459	0.82687	0.00556	2.1771
4	37-39	0.0178	0.01167	0.17355	0.50890	0.00462	0.78381	0.00590	2.5400
4	40-42	0.0234	0.01538	0.18893	0.40020	0.00360	0.77519	0.00464	2.1771
4	43-45	0.0245	0.01610	0.20503	0.40250	0.00442	0.94746	0.00467	0.8467
4	46-48	0.0186	0.01222	0.21725	0.36670	0.00385	0.90439	0.00425	0.9728
4	49-51	0.0195	0.01279	0.23004	0.38490	0.00258	0.57709	0.00446	11.4300
4	52-54	0.0219	0.01435	0.24439	0.49910	0.00543	0.93885	0.00579	11.4300
4	55-57	0.0288	0.01892	0.26331	0.27370	0.00238	0.74935	0.00317	11.4300
4	58-60	0.0182	0.01194	0.27525	0.34810	0.00365	0.90439	0.00404	11.4300
4	61-63	0.0151	0.00993	0.28518	0.38120	0.00255	0.57709	0.00442	1.2032
4	64-66	0.0148	0.00970	0.29488	0.38510	0.00419	0.93885	0.00447	0.9728
4	67-69	0.0204	0.01339	0.30827	0.31560	0.00274	0.74935	0.00366	1.5240
4	70-72	0.0347	0.02275	0.33102	0.40240	0.00422	0.90439	0.00467	1.6329
4	73-75	0.0550	0.03605	0.36707	0.55040	0.00544	0.85271	0.00638	0.1725

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Time of sample collection	Glass runoff, volume (mL)	Glass runoff, field pH	Glass runoff, laboratory pH	Glass runoff, field conductivity (µS/cm)	Glass runoff, alkalinity (meq/L)	Glass runoff, chloride (meq/L)	Glass runoff, nitrate (meq/L)	Glass runoff, sulfate (meq/L)
5	1-3	346	112	4.69	4.71	11.0	-0.01950	0.00530	0.02023	0.04665
5	4-6	352	114	4.72	4.56	7.1	-0.02910	0.00311	0.00650	0.02724
5	7-9	355	114	4.79	4.64	6.1	-0.02430	0.00909	0.00302	0.03600
5	10-12	359	94	4.69	4.54	7.5	-0.03060	0.01014	0.00380	0.02640
5	13-15	409	106	4.16	4.11	23.0	-0.08230	0.00380	0.01627	0.07621
5	16-18	717	114	3.98	3.94	34.2	-0.12350	0.00649	0.03829	0.11574
5	19-21	728	89	4.34	4.30	13.7	-0.05280	0.00248	0.01361	0.04281
5	22-24	731	62	4.74	4.60	5.4	-0.02640	0.00180	0.00071	0.01941
5	25-27	739	90	4.89	4.68	4.6	-0.02190	0.00241	0.00150	0.01564
5	28-30	749	92	4.83	4.66	4.9	-0.02290	0.00180	0.00059	0.01593
5	31-33	804	88	4.74	4.58	6.0	-0.02770	0.00180	0.00114	0.01900
5	34-35	1017	55	4.06	4.01	31.4	-0.10400	0.00455	0.03639	0.10281
6	1-3	1716	112	3.88	3.95	49.2	-0.14400	0.01238	0.06291	0.15558
6	4-6	1717	114	4.08	4.14	29.7	-0.09340	0.00694	0.03349	0.08211
6	7-9	1718	114	4.33	4.39	16.3	-0.05110	0.00446	0.01807	0.04878
6	10-12	1719	94	4.41	4.51	12.6	-0.03890	0.00338	0.01259	0.03498
6	13-15	1719.5	106	4.55	4.58	10.2	-0.03290	0.00274	0.00936	0.02915
6	16-18	1720	114	4.74	.4.76	6.7	-0.02140	0.00895	0.00572	0.01974
6	19-21	1721	89	4.59	4.62	9.4	-0.02980	0.00262	0.00853	0.02465
6	22-24	1722	62	4.51	4.55	11.0	-0.03520	0.00349	0.01078	0.02838
6	25-27	1730	62	4.24	4.29	20.6	-0.06410	0.00384	0.02406	0.04918
6	28-30	1738	104	4.13	4.18	25.4	-0.08410	0.00521	0.02890	0.06254
6	31-33	1742	77	4.38	4.43	13.7	-0.04520	0.00278	0.01362	0.03347
6	34-36	1753	65	4.35	4.39	15.8	-0.05060	0.00301	0.02098	0.03853
6	37-39	2041	65	4.10	4.15	27.0	-0.90140	0.00479	0.04309	0.06055
6	40-42	2050	60	4.23	4.28	20.1	-0.06880	0.00387	0.02420	0.05156
6	43-45	2053	77	4.31	4.35	17.4	-0.05770	0.00291	0.01805	0.05089
6	46-48	2057	81	4.38	4.41	15.0	-0.04970	0.00910	0.01576	0.04370
6	49-51	2104	78	4.55	4.57	9.5	-0.03340	0.00180	0.00863	0.02836
6	52-54	2110	93	4.62	4.68	7.2	-0.02630	0.00180	0.00451	0.02305
6	55-57	2113	85	4.67	4.68	8.9	-0.02600	0.00180	0.00388	0.02226
6	58-60	2114	92	4.77	4.78	6.0	-0.02050	0.00180	0.00144	0.01760
6	61-63	2116	86	4.76	4.76	5.8	-0.02160	0.00180	0.00199	0.01837
6	64-65	2126	77	4.26	4.27	20.2	-0.06730	0.00378	0.03253	0.05154
7	1-3	210	95	3.34	3.37	203.0	0	0.02393	0.26006	0.43308
7	4-6	328	83	3.45	3.48	157.0	-0.40140	0.01711	0.21110	0.29717
7	7-9	355	NA	NA	NA	NA	NA	NA	NA	NA
7	10-12	419	91	3.67	3.69	95.8	-0.23580	0.01085	0.11545	0.17872
7	13-15	457	86	3.78	3.81	74.9	-0.18280	0.00911	0.08821	0.14439
7	16-18	523	65	3.92	4.08	39.2	-0.09780	0.00456	0.03320	0.08096
7	19-21	541	78	4.21	4.24	17.4	-0.06680	0.00283	0.01406	0.05946
7	22-25	920	73	4.25	4.33	22.3	-0.05280	0.00244	0.00870	0.05013

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Glass runoff, calcium (meq/L)	Glass runoff, magnesium (meq/L)	Glass runoff, sodium (meq/L)	Stone runoff, volume (mL)	Stone runoff, field pH	Stone runoff, laboratory pH	Stone runoff, field conductivity (µS/cm)
5	1-3	0.02590	0.00640	0.0098	108	7.26	6.84	25.8
5	4-6	0.00850	0.00210	0.0033	106	7.33	6.76	20.7
5	7-9	0.00520	0.00110	0.0018	103	7.26	6.67	17.5
5	10-12	0.00500	0.00090	0.,0017	114	7.27	6.69	15.8
5	13-15	0.00950	0.00180	0.0024	103	7.27	6.83	21.6
5	16-18	0.02230	0.00520	0.0045	90	7.51	7.09	41.0
5	19-21	0.00510	0.00100	0.0026	114	7.51	6.73	23.1
5	22-24	0.00220	0.00050	0.0017	89	7.32	6.72	12.2
5	25-27	0.00140	0.00040	0.0018	114	7.34	6.86	17.5
5	28-30	0.00120	0.00030	0.0011	114	7.41	6.90	18.5
5	31-33	0.00120	0.00030	0.0008	114	7.48	6.96	20.7
5	34-35	0.01660	0.00370	0.0068	70	6.61	7.15	38.4
6	1-3	0.01920	0.00530	0.0258	71	7.15	6.75	25.1
6	4-6	0.00730	0.00180	0.0084	103	7.00	6.63	21.7
6	7-9	0.00800	0.00120	0.0066	114	7.05	6.71	23.3
6	10-12	0.00570	0.00070	0.0033	114	7.08	6.70	20.1
6	13-15	0.00560	0.00050	0.0021	114	7.03	6.68	16.6
6	16-18	0.00370	0.00040	0.0014	114	7.13	6.68	12.5
6	19-21	0.00370	0.00050	0.0015	114	7.10	6.75	11.0
6	22-24	0.00350	0.00050	0.0015	114	7.13	6.83	13.3
6	25-27	0.00580	0.00090	0.0033	114	7.23	7.06	21.3
6	28-30	0.00430	0.00080	0.0025	114	7.25	6.98	20.2
6	31-33	0.00190	0.00050	0.0014	114	7.33	6.99	15.1
6	34-36	0.00430	0.00090	0.0032	113	7.48	7.20	22.9
6	37-39	0.00640	0.00120	0.0024	114	7.42	6.91	26.5
6	40-42	0.00330	0.00080	0.0014	97	4.43	6.94	24.0
6	43-45	0.00270	0.00060	0.0015	108	7.31	6.76	16.0
6	46-48	0.00230	0.00050	0.0007	114	7.30	6.78	15.2
6	49-51	0.00140	0.00040	0.0009	114	7.32	6.82	15.0
6	52-54	0.00100	0.00030	0.0009	114	7.34	6.85	14.4
6	55-57	0.00120	0.00030	0.0012	114	7.23	6.80	11.4
6	58-60	0.00100	0.00020	0.0008	80	7.24	6.73	9.0
6	61-63	0.00120	0.00030	0.0007	111	7.25	6.79	10.4
6	64-65	0.00900	0.00170	0.0019	77	7.54	7.26	30.6
7	1-3	0.10270	0.01860	0.01420	92	7.43	7.43	113.0

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Stone runoff, alkalinity (meq/L)	Stone runoff, chloride (meq/L)	Stone runoff, nitrate (meq/L)	Stone runoff, sulfate (meq/L)	Stone runoff, calcium (meq/L)	Stone runoff, magnesium (meq/L)	Stone runoff, sodium (meq/L)
5	1-3	0.27890	0.00571	0.02520	0.08468	0.36870	0.02030	0.0054
5	4-6	0.19810	0.00292	0.01331	0.09100	0.29070	0.01630	0.0016
5	7-9	0.14370	0.00537	0.00749	0.09412	0.23200	0.01120	0.0027
5	10-12	0.15890	0.00180	0.00590	0.06544	0.22170	0.00920	0.0011
5	13-15	0.21560	0.00301	0.01429	0.07956	0.30070	0.00990	0.0013
5	16-18	0.43570	0.00648	0.03766	0.12561	0.57760	0.02270	0.0023
5	19-21	0.28130	0.00342	0.01664	0.05333	0.34040	0.01250	0.0011
5	22-24	0.15100	NA	0.00199	0.02625	0.17650	0.00510	0.0008
5	25-27	0.21540	0.00695	0.00184	0.02432	0.23770	0.00770	0.0009
5	28-30	0.25210	0.00882	0.00208	0.02265	0.27460	0.00670	0.0005
5	31-33	0.28100	NA	0.00205	0.02482	0.30380	0.00660	0.0003
5	34-35	0.46100	0.00392	0.01698	0.06927	0.52560	0.01850	0.0015
6	1-3	0.12570	0.01235	0.05011	0.14288	0.29270	0.01300	0.0108
6	4-6	0.08570	0.00738	0.04618	0.13808	0.22400	0.03330	0.0065
6	7-9	0.09810	0.00502	0.02495	0.16529	0.26120	0.01950	0.0046
6	10-12	0.09000	0.00468	0.02304	0.14228	0.22720	0.02380	0.0026
6	13-15	0.08690	0.00394	0.01323	0.12109	0.20280	0.01150	0.0023
6	16-18	0.08190	0.00240	0.00755	0.06978	0.14990	0.00740	0.0008
6	19-21	0.08920	0.00245	0.00880	0.04422	0.13610	0.00530	0.0012
6	22-24	0.11420	0.00350	0.01215	0.04297	0.16150	0.00650	0.0016
6	25-27	0.20430	0.00413	0.02430	0.06251	0.27940	0.00950	0.0022
6	28-30	0.16810	0.00422	0.02872	0.06728	0.25100	0.00720	0.0019
6	31-33	0.16330	0.00323	0.01388	0.03796	0.20550	0.00650	0.0014
6	34-36	0.26740	0.00297	0.01565	0.04316	0.31390	0.01030	0.0013
6	37-39	0.24980	0.00440	0.04295	0.07009	0.35020	0.01160	0.0015
6	40-42	0.25140	0.00312	0.02644	0.05763	0.31400	0.01050	0.0012
6	43-45	0.14440	0.00238	0.01882	0.05434	0.20350	0.00540	0.0014
6	46-48	0.14760	0.00217	0.01725	0.04762	0.19760	0.00630	0.0010
6	49-51	0.16310	0.00542	0.01016	0.03177	0.19550	0.00490	0.0011
6	52-54	0.16650	0.00180	0.00584	0.02641	0.19270	0.00510	0.0008
6	55-57	0.12530	0.00180	0.00460	0.02506	0.14930	0.00400	0.0010
6	58-60	0.09610	0.00180	0.00227	0.01952	0.11520	0.00280	0.0016
6	61-63	0.11750	0.00180	0.00208	0.01968	0.13520	0.00310	0.0009
6	64-65	0.37620	0.00261	0.01525	0.04544	0.41890	0.01030	0.0009
7	1-3	0.55090	0.02271	0.24475	0.38467	1,16400	0.04310	0.00630
7	4-6	0.51770	0.01956	0.17032	0.26130	0.94450	0.04610	0.00720
7	7-9	0.57090	0.02250	0.22894	0.32154	1.08800	0.05290	0.00710
7	10-12	0.46220	0.01014	0.11857	0.18933	0.74450	0.03600	0.00560
7	13-15	0.56750	0.00715	0.07481	0.13122	0.76880	0.02950	0.00470
7	16-18	0.57770	0.00541	0.04196	0.09943	0.70950	0.02870	0.00350
7	19-21	0.57120	0.00401	0.02245	0.07516	0.66710	0.02340	0.00270
7	22-25	0.53420	0.00361	0.01681	0.07177	0.63720	0.02520	0.00320

APPENDIX. Sequential runoff data from glass and carbonate-stone microcatchments during seven summer storms in the Adirondack Mountains, New York, 1987–89--Continued

Storm[1]	Sequential sample number	Glass runoff, hydrogen ion (meq/L)	Glass runoff, hydrogen ion load (meq/m²)	Cumulative hydrogen ion loading (meq/m²)	Net calcium (meq/L)	Recession (µm)	Rain depth (mm)	Micrometer recession per millimeter rain	Rain rate (mm/hr)
5	1-3	0.0204	0.00837	0.00837	0.34280	0.00370	0.58127	0.00636	2.8575
5	4-6	0.0191	0.00781	0.01618	0.28220	0.00299	0.57051	0.00524	7.6200
5	7-9	0.0162	0.00665	0.02283	0.22680	0.00233	0.55436	0.00421	15.2400
5	10-12	0.0204	0.00837	0.03120	0.21670	0.00247	0.61356	0.00402	11.4300
5	13-15	0.0692	0.02837	0.05957	0.29120	0.00300	0.55436	0.00540	4.5720
5	16-18	0.1047	0.04293	0.10250	0.55530	0.00499	0.48439	0.01030	0.2432
5	19-21	0.0457	0.01874	0.12124	0.33530	0.00382	0.61356	0.00622	4.1564
5	22-24	0.0182	0.00746	0.12870	0.17430	0.00155	0.47901	0.00323	15.2400
5	25-27	0.0129	0.00528	0.13398	0.23630	0.00269	0.61356	0.00438	5.7150
5	28-30	0.0148	0.00606	0.14005	0.27340	0.00311	0.61356	0.00507	4.5720
5	31-33	0.0182	0.00746	0.14751	0.30260	0.00345	0.61356	0.00561	3.0480
5	34-35	0.0871	0.03571	0.18322	0.50900	0.00356	0.37675	0.00944	0.3438
6	1-3	0.1318	0.08648	0.08468	0.27350	0.00194	0.61154	0.00317	22.8600
6	4-6	0.0832	0.05456	0.14104	0.21670	0.00223	0.88717	0.00251	45.7200
6	7-9	0.0468	0.03068	0.17172	0.25320	0.00288	0.98191	0.00294	45.7200
6	10-12	0.0389	0.02552	0.19725	0.22150	0.00252	0.98191	0.00257	45.7200
6	13-15	0.0282	0.01849	0.21573	0.19720	0.00225	0.98191	0.00229	91.4400
6	16-18	0.0182	0.01194	0.22767	0.14620	0.00166	0.98191	0.00170	91.4400
6	19-21	0.0257	0.01686	0.24453	0.13240	0.00151	0.98191	0.00154	45.7200
6	22-24	0.0309	0.02027	0.26481	0.15800	0.00180	0.98191	0.00183	45.7200
6	25-27	0.0575	0.03775	0.30255	0.27360	0.00311	0.98191	0.00317	5.7150
6	28-30	0.0741	0.04863	0.35118	0.24670	0.00281	0.98191	0.00286	5.7150
6	31-33	0.0417	0.02735	0.37853	0.20360	0.00232	0.98191	0.00236	11.4300
6	34-36	0.0447	0.02930	0.40783	0.30960	0.00349	0.97330	0.00359	4.1564
6	37-39	0.0794	0.05211	0.45994	0.34380	0.00391	0.98191	0.00399	0.2721
6	40-42	0.0589	0.03863	0.49857	0.31070	0.00301	0.83549	0.00360	5.0800
6	43-45	0.0490	0.03213	0.53070	0.20080	0.00217	0.93023	0.00233	15.2400
6	46-48	0.0417	0.02735	0.55805	0.19530	0.00222	0.98191	0.00226	11.4300
6	49-51	0.0282	0.01849	0.57653	0.19410	0.00221	0198191	0.00225	6.5314
6	52-54	0.0240	0.01574	0.59227	0.19170	0.00218	0.98191	0.00222	7.6200
6	55-57	0.0214	0.01403	0.60630	0.14810	0.00169	0.98191	0.00172	15.2400
6	58-60	0.0170	0.01114	0.61744	0.11420	0.00091	0.68906	0.00132	45.7200
6	61-63	0.0174	0.01140	0.62884	0.13400	0.00149	0.95607	0.00155	22.8600
6	64-65	0.0550	0.03605	0.66489	0.40990	0.00315	0.66322	0.00475	4.5720
7	1-3	0.4571	0.29985	0.29985	1.06130	0.00970	0.78854	0.01231	0.0000
7	4-6	0.3548	0.23276	0.53261	0.89490	0.00520	0.50129	0.01038	0.5862
7	7-9	NA	–	0.53261	1.08800	0.00608	0.48191	0.01262	1.6933
7	10-12	0.2138	0.14025	0.67286	0.72590	0.00432	0.51378	0.00842	1.9050
7	13-15	0.1660	0.10887	0.78173	0.75440	0.00543	0.62059	0.00875	1.2032
7	16-18	0.1202	0.07887	0.86060	0.70330	0.00377	0.46210	0.00815	1.7585
7	19-21	0.0617	0.04045	0.90104	0.66280	0.00414	0.53876	0.00768	2.5400
7	22-25	0.0562	0.03689	0.93793	0.63290	0.00591	0.80491	0.00734	0.2088

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[1] Glass and marble runoff volumes for Storms 1 and 2 were calculated from one microcatchment area.

[2] Net calcium calculated by subtracting calcium concentrations in glass runoff from calcium concentration in storm runoff.