Atmospheric Environment Vol 26A No 11 pp 2001-2017 1992
Printed in Great Britain

0004 6981 92 $5 00+0 00

REGIONAL ANALYSIS OF CLOUD CHEMISTRY AT HIGH ELEVATIONS IN THE EASTERN UNITED STATES

ZHENG LI and VINEY P ANEJA
Department of Marine, Earth and Atmospheric Sciences, North Carolina State University Raleigh NC 27695-8208 U S A

(*First received* 15 *February* 1991 *and in final form* 13 *January* 1992)

Abstract—Results from the collection and chemical analysis of cloudwater samples collected from May to October 1986–1988 from the five high-elevation ($\gtrsim$950 m MSL) Mountain Cloud Chemistry Program (MCCP) sites (Whiteface Mountain, NY, Mt Moosilauke, NH, Shenandoah Park, VA, Whitetop Mountain, VA, Mt Mitchell, NC) in the eastern United States are summarized The resulting database documents the regional chemical climatology of high-elevation forest ecosystems in the eastern U S Clouds occurred at these sites on 32–77% of the days during the sample collection period More than 90% of cloud samples were acidic (pH < 5 0) The lowest cloudwater pH (2 29 integrated 1-h collection period) was recorded at Mt Mitchell, NC At all sites sulfate and nitrate were the dominant anions and hydrogen and ammonium were the dominant cations in cloudwater samples Mount Mitchell received the most acidic clouds and highest chemical exposures, while the Whiteface summit site received the least acidic and lowest chemical exposures compared to other MCCP high-elevation sites Cloud pH and major chemical components exhibited a seasonal trend with the maxima during the summer months, and correlated well with temperature and ozone concentrations The mean equivalent ratios of SO_4^{2-} to NO_3^- were found to be 1 9–3 9 at these sites It is noted that SO_4^{2-} correlated highly with hydrogen ion, suggesting that contribution to cloud acidity by sulfate and/or its precursors may be significant

Key word index Cloud chemistry, regional analysis, ozone, chemical exposure

1 INTRODUCTION

High-elevation forests in the eastern United States have shown signs of injury and decline during the past two decades In recent years, there has been increasing concern with the possible impact of atmospheric acidity on forested ecosystems (Klein and Perkins, 1988, Cowling, 1989, Bruck *et al* 1989, Jacobson *et al*, 1990b, Hertel *et al*, 1990) It is now believed that acidic cloud deposition may contribute to observed forest decline at high-elevation locations where mountain slopes are frequently immersed in clouds (Jacobson *et al*, 1990a, Saxena and Lin, 1990, Aneja *et al*, 1990a, 1992, Cowling *et al*, 1991)

Acidic clouds and fogs have been characterized over several decades in the U S and Europe, both in urban and rural areas Table 1 shows examples of cloud acidity measurements at some of those locations The cloud pH values reported by these investigators ranged from 2 2 to 7 6 Most authors focused on individual case studies, except for Weathers *et al* (1986), who studied a widespread acid cloud event at six non-urban sites in the eastern U S However, their study was based on one single event and limited to moderate elevation No research has been done for high-elevation regional cloud chemistry climatological analysis from north to south in the eastern U S to compare the acidity exposures between different sites Possible forest decline in high-elevation ecosystems across the eastern U S necessitated documentation of the chemical exposure and distribution of clouds at high elevation on a regional scale

Beginning in 1986, a series of measurements, including major cations and anions in cloud water and precipitation, gas-phase measurements of ozone, sulfur dioxide and nitrogen oxides, and meteorological parameters (Aneja *et al*, 1992) were made at six remote sites in the eastern U S These measurements were made as part of the Mountain Cloud Chemistry Project (MCCP) sponsored by the U S Environmental Protection Agency Five high-elevation ($\gtrsim$ 950 m MSL) sites were selected from 35 to 45°N to be representatives of the geographic and meteorological variability in this large region One low-elevation site (Howland, ME, 65 m MSL) was instrumented to allow evaluation of the impact of elevational gradient In these ecosystems, red spruce (*Picea rubens* Sarg) and Fraser fir (*Abies fraseri* [Pursh] Poir) are the dominant tree species, they have shown signs of decline above the cloud base, which is frequently observed around 800–1200 m (Mohnen *et al*, 1990a)

The objectives of this research are to (i) characterize the exposure of montane forested ecosystems to chemicals in cloud water at high elevations in the eastern U S, (ii) determine north–south gradients of cloud chemistry based on observations made at five high-elevation MCCP sites, (iii) study the regional chemical climatology at high elevations in the eastern U S

Table 1 The ranges of cloudwater acidity observed at other locations

Reference	Year of study	Location	Type of collector	Range of pH
Cloud water reported from low elevation				
Houghton (1955)	1954	Northeast U S A	stainless steel or nickel screen	4 5–7 2
Mrose (1966)	1957	Germany	previous cloth	3 8–5 1
Lazrus *et al* (1970)	1967	Puerto Rico	aluminum screen	4 9–5 4
Waldman *et al* (1982)	1981	California, U S A	Caltech rotating arm	2 2–4 0
Munger *et al* (1983)	1981–1982	California, U S A	Caltech Teflon strings	2 2–5 8
Fuzzi *et al* (1984)	1982	New York, U S A	screen impactor	4 3–6 4
Jacob *et al* (1985)	1982–1983	California, U S A	Caltech rotating arm	2 2–6 3
Muir *et al* (1986)	1985–1986	Midwestern U S A	Caltech rotating arm	2 9–4 1
Weathers *et al* (1986)	1984	Eastern U S A	Teflon strings	2 9–3 0
Cloud water collected by aircraft				
Oddie (1962)	1960	United kingdom	glass tube	4 4–7 2
Petrenchunk and Drozdova (1966)	1961–1964	U S S R	integrated sample	3 4–5 9
Scott (1978)	1976	Australia	centrifuge	4 6–7 5
Scott and Laulainen (1979)	1977	Michigan, U S A	nylon wand	3 7–4 0
Daum *et al* (1984)	1981–1983	Eastern U S A	slotted-rod impactor	3 1–6 1
Saxena *et al* (1985)	1982–1983	McMurdo, Antarctica	Teflon probe	4 9–6 2
Khemani *et al* (1987)	1983–1985	Pune, India	stainless steel sheets	6 3–7 6
Hegg and Hobbs (1981)	1979	Northwestern U S A	centrifuge	4 2–6 5
Cloud water collected at mountain sites				
Okita (1968)	1963	Japan	copper screen	3 5–6 5
Castillo (1979)	1976	Whiteface Mtn, NY, U S A	rotating stainless steel tube	3 4–4 2
Falconer and Falconer (1980)	1977–1979	Whiteface Mtn, NY, U S A	ASRC, Teflon string	2 7–4 7
Weathers *et al* (1986)	1984	Eastern U S A	Teflon strings	2 8–3 1
Mohnen and Kadlecek (1989)	1982–1987	Whiteface Mtn, NY, U S A	ASRC, Teflon string	2 5–4 8
Aneja *et al* (1990a)	1987	Mt Mitchell, NC, U S A	Caltech, Teflon string	2 9–4 6
Aneja *et al* (1990b)	1986–1988	Mt Mitchell, NC, U S A	ASRC, Teflon string	2 2–5 5
Saxena and Lin (1990)	1986–1987	Mt Mitchell, NC, U S A	ASRC, Teflon string	2 2–5 4