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Na^+/Cl^- ratios in rain across the USA, 1982–1986

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ABSTRACT

Ratios of Na^+/Cl^- for annual rain collections for 178 sites in the NADP/NTN network across the USA were compared to the same ratio for sea water. 48% of the sites had ratios which were, within experimental error, the same as sea water. 32% of the sites, all in western states, had Na^+/Cl^- ratios greater than sea water. These higher ratios are attributed to Na^+ from feldspar and clay aerosols. The remainder (20%) of the sites, with ratios less than sea water are mostly in a contiguous area composed of lower Michigan, lower Wisconsin, eastern Illinois, Indiana, Ohio, Kentucky, West Virginia, northern Virginia, Pennsylvania, New York and Massachusetts. This area coincides with the areas of highest SO_4^{2-} and NO_3^- concentrations in rain and is believed to receive imputs of Cl^- from coal-fired power plants.

1. Introduction

Sea salt is an ubiquitous component of tropospheric aerosols. With NaCl the major component of sea salt the Na^+/Cl^- ratio of sea salt would be expected for rain water without other sources for these ions. Generally, the Na^+/Cl^- ratio of rain is near the sea salt equivalents ratio of 0.86 (Junge, 1963). However, perturbations from this number have been observed.

In earlier work, Junge and Werby (1958) showed an increase in the Na^+/Cl^- ratio in rain as the collection site was moved inland from the ocean. This increase was attributed to mineral aerosols from land sources which supplied leachable Na^+ but no Cl^-. Soils and rocks have only minor amounts of Cl^- (Mason, 1966). Since then several authors (Erikson, 1959, 1960; Hitchcock et al., 1980; Charlson et al., 1978; and Martens et al., 1973) have proposed the loss of Cl^- from NaCl aerosols as another method for increasing the Na^+/Cl^- in rain. These latter schemes involved the loss of Cl^-, particularly at coastal sites, by reaction of the NaCl aerosol with atmospheric acids or ozone to liberate HCl or Cl_2.

A large data base is now available from the network of sites across the USA which are part of the National Atmospheric Deposition Program (NADP)/National Trends Network (NTN). It seemed appropriate to reexamine the above findings with these more recent data which more intensively cover a larger area for a longer time. Improvements in collecting, handling, and chemically analyzing the rain samples have undoubtedly occurred due to the emphasis placed on these aspects by operator training, improved equipment, monitoring and quality assurance programs. In the present paper the Na^+/Cl^- annual flux ratios of rain collections for most of the sites of the NADP/NTN have been examined over a 5-year period, 1982–1986. The data presented are from 178 sites in 46 states.

2. Methods

Analyses of rains from the NADP/NTN network of sites have been summarized by year for 1982, 1983, 1984, 1985, and 1986 (NADP, 1985; NADP/NTN, 1985, 1986, 1987a, 1987b). Only those sites and collections passing NADP/NTN criteria for site location, sample handling and measurement protocol are included in the annual summaries. The NADP/NTN criteria are several but two of the more important are: (a) only "wet only" samples, not bulk collected

samples, are included and (b) only sites with valid samples for at least 75% of the total precipitation at the site are included. Overall the valid samples on which data are reported here average 91% of the total precipitation per site.

All chemical analyses were made at the Central Analytical Laboratory of the Illinois State Water Survey. The analytical methods and detection limits have been summarized by Peden (1986) and the quality assurance program by Lockard (1987). Na^+ was analyzed by flame atomic absorption and Cl^- by automated colorimetric ferricyanide (1982–83) and by ion chromatography (1984–86). Bias and Relative Standard Deviation (RSD) were determined over the years on Na^+ and Cl^- analyses using samples representing the 25th and 75th percentile concentrations. The Bias for both Na^+ and Cl^- ranged from 0.0–4.0%. RSD ranged from 0.6–3.0% for Na^+ and for Cl^- 3.5–4.7% (1982–83) and 1.6–2.3% (1983–86). Bias and RSD are defined as follows:

$$\% \text{ Bias} = 100(V_m - V_t)/V_t,$$

where V_m = measured value, V_t = true value;

$$\text{RSD} = 100S/\overline{X},$$

where S = standard deviation, $\overline{X}$ = mean value.

3. Results and discussion

The yearly fluxes of Na^+ and Cl^- for rains were determined for the various sites for the years 1982–1986. Flux units of gram equivalents per hectare per year were used which gives for flux ratios the units of equivalents per equivalent. The Na^+/Cl^- flux ratios were compared to the concentration ratio, in equivalents, for sea water (SW) via the ratio $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ where $(Na^+/Cl^-)_{SW} = 0.86$. Fig. 1 gives the areal plot of these wet-deposition flux ratios $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ for the various sites in the USA for 1982–1986. In this plot, there are 178 total sites with the data representing only 1 year in the 1982–1986 period at 34 sites, the average of two years at 54 sites, three years at 30 sites, four years at 22 sites and five years at 38 sites. Thus, the plotted value is the average value representing the 1–5 year operating lifetimes of the sites.

48% of the values in Fig. 1 are in the range 0.9–1.1 and are mostly in the unshaded areas of the coastal, midcontinent and southeastern states. Those values outside this range are in the shaded areas. Those exceeding 1.1 and representing 32% of the sites are in area A. This area is characterized by a Na^+ contribution from land augmenting the sea-salt contribution. The Na^+ is believed to come from clay and feldspar, $(NaK\,Ca/2)AlSi_3O_8$, aerosols in the troposphere which are scavenged by rain and reacted via ion exchange or acid leaching. Clay and feldspar probably originated in the Western Plains and were transported eastwardly by winds. In support of this interpretation, we have identified both clay and feldspar by X-ray diffraction in the particulate matter of the wet-deposition of the Fayetteville, Arkansas, site (Wagner and Steele, 1986) and 21 other western USA sites in Fig. 1. From the diffractograms, 1–5 times the requisite feldspar in the form of andesine (8% Na) was potentially available to provide the excess Na^+, i.e., that greater than 0.86 Cl^-. The principal clay was illite (0.4% Na) which would be a minor source of Na^+ but a good source of K^+.

Other facts support feldspar aerosol as a source of the excess Na^+. In various laboratory experiments we have observed the reaction of clays and feldspars with mineral acids. Others have identified clays and feldspars in western aerosols (Davis et al., 1982; Smith and Twiss, 1965; Smith et al., 1970). The deposition rates of aerosol dusts decrease from west to east (Applin and Jersak, 1986). Historically, dust storms have moved from west to east. Clay and feldspar are staples of the eolian deposits of loess in the midwest (Pettijohn, 1957). Other cations expected from terrestrial sources, Ca^{2+} and K^+, have their highest concentration isopleths for wet deposition centered in the western states (NADP/NTN, 1987b).

Area A is characterized by generally higher values of the $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratios in its western part which decrease to 1.0 to the east with area A entering in an interfingering way the corridor separating areas A and B. This regional behavior has four exceptions with ratios less than 1.0 and these show through the shaded area A in Fig. 1. This situation indicates that the regional condition of high content of feldspar and clay aerosols can be overridden by local conditions.

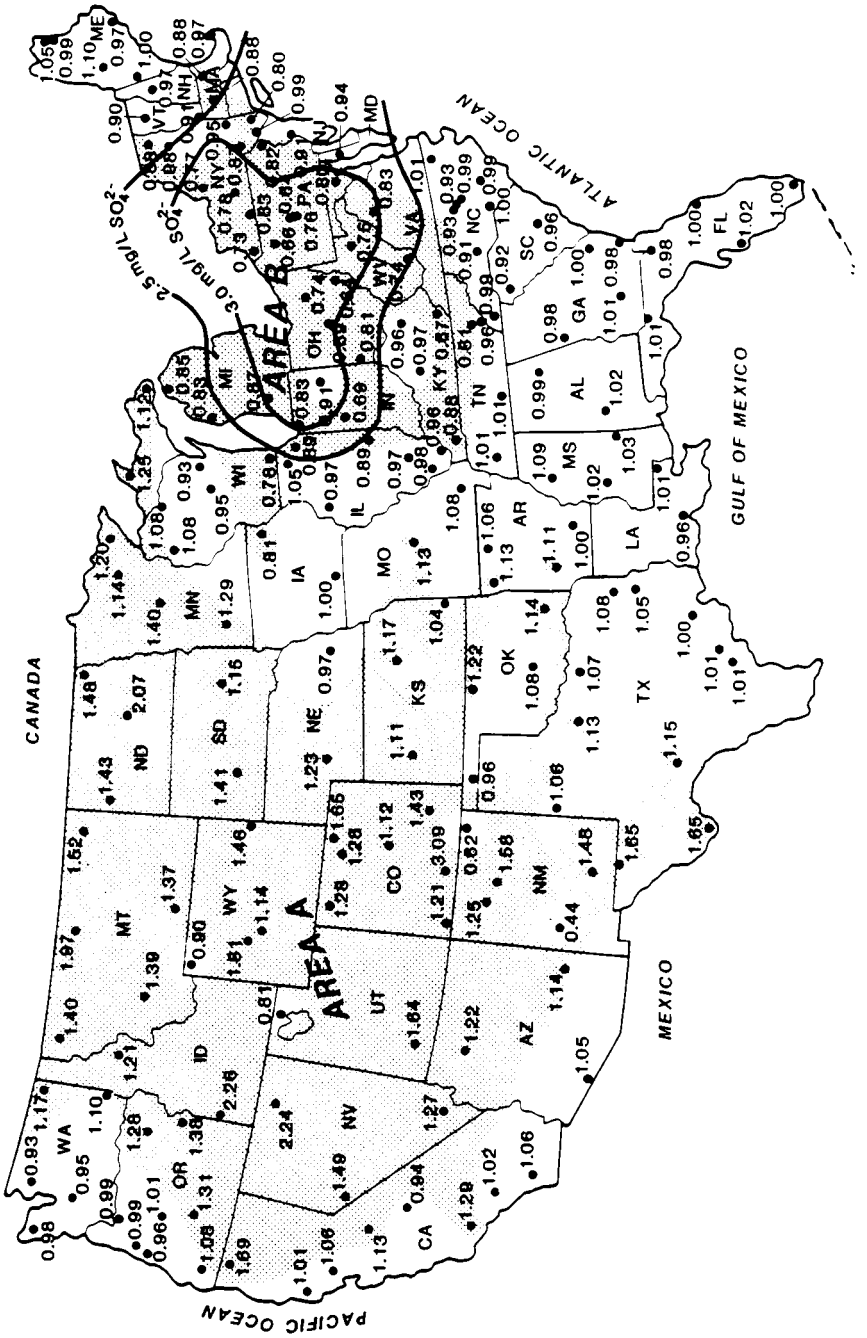

Fig. 1. $(Na^+/Cl^-)/(Na^+/Cl^-)_{sw}$ ratios in the wet deposition of various collection sites in the NADP/NTN network for 1982–1986.

The local conditions are unknown but are thought to be the same as those existing in area B, i.e., high Cl^- output of nearby coal-burning power plants.

A surprising facet of these data is the gain of Cl^- relative to Na^+ in shaded area B in Fig. 1. This area includes Michigan (MI), eastern Illinois (IL), Indiana (IN), Ohio (OH), southern Wisconsin (WI), the northeastern tip of Iowa (IA), Kentucky (KY), West Virginia (WV), Pennsylvania (PA), western New York (NY), eastern Massachussets (MA), and northern Virginia (VA). In this area the $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratio is less than 0.90 and is attributed to HCl from coal burning power plants. Area B is coincident with the area of highest SO_4^{2-} and NO_3^- concentrations in wet-deposition in the USA (NADP/NTN, 1985, 1986, 1987a, 1987b) and this has commonly been attributed to coal burning. The highest concentration isoplets for SO_4^{2-} for 1986 are shown in Fig. 1. The highest concentration isopleths for NO_3^- are approximately the same. Furthermore, the core states of area B—Ohio, Pennsylvania, Indiana and Illinois—are the four highest states in emission of SO_2, 2.7–1.5 million tons/year in 1980 (Gilleland et al., 1985).

For orientation purposes the cores of areas A and B have precipitation-weighted mean chloride ion concentrations (mg/l) of about 0.08 and 0.13 respectively. Along the Atlantic, Pacific, and Gulf coasts the corresponding figure is 0.40 which falls to 0.20 within about 150 km inland. The corresponding annual fluxes for the cores of areas A and B are respectively 30 and 120 $mg/m^2/yr$. These numbers are for 1986 when the areal sampling was the most extensive.

It is of interest to know what cation accompanies the excess Cl^- in area B. Table 1 gives the fluxes of the various cations for 9 sites in area B at which a significant reduction in the $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratio occurred. At 6 sites, the reduction is accompanied by an increase in H^+ flux, a slight decrease in H^+ at 2 sites, and a significant decrease at 1 site. The Cl^- flux increased, as expected, at 6 sites but decreased at 3 sites as the Na^+/Cl^- ratio decreased. The H^+/Cl^- flux ratio increased as the Na^+/Cl^- flux ratio decreased at only 4 sites and decreased at 5 sites. Possible complications to this type of comparison

Table 1. *Change in cation flux with lowering of Na^+/Cl^- ratio (eq./eq.)**

Site	Year	Na^+/Cl^- / $(Na^+/Cl^-)_{SW}$	Annual flux, milleq./m^2/yr Ca^{2+}	Mg^{2+}	K^+	Na^+	NH_4^+	H^+	Unknown Cl^-	H^+/Cl^-
1. Indiana, Indiana Dunes N.P.	1983	0.90	20	6.2	0.87	3.3	22	37	0.3	4.2	8.8
Indiana, Indiana Dunes N.P.	1984	0.74	15	5.0	0.74	3.1	17	36	0.3	4.8	7.5
2. Indiana, Purdue Agri Farm	1984	0.93	10	4.0	0.95	3.8	27	48	1.7	4.8	9.9
Indiana, Purdue Agri Farm	1983	0.24	11	3.0	0.56	2.5	17	45	0.1	12	3.9
3. Michigan, Douglas Lake	1986	1.03	6.5	2.1	0.36	1.4	11	24	5.4	1.7	14
Michigan, Douglas Lake	1985	0.78	11	3.8	0.66	2.1	19	36	3.5	3.1	12
4. Michigan, Kellogg Bio. Station	1982	0.86	11	4.2	0.61	2.1	18	42	4.2	2.8	15
Michigan, Kellogg Bio. Station	1985	0.74	11	3.8	0.61	2.9	26	56	6.8	4.5	12
5. New York, Bennett Bridge	1984	0.91	12	4.3	1.1	4.6	25	70	1.2	5.9	12
New York, Bennett Bridge	1985	0.74	10	3.5	0.92	4.1	28	86	3.7	6.5	13
6. Pennsylvania, Leading Ridge	1982	1.01	9.5	3.2	0.92	5.1	20	72	7.8	5.9	12
Pennsylvania, Leading Ridge	1985	0.65	6.0	2.5	0.82	3.17	14	65	5.2	5.6	11
7. Tennessee, Walker Branch	1984	0.87	11	4.0	1.3	7.0	19	53	1.3	9.3	5.7
Tennessee, Walker Branch	1985	0.62	7	2.8	0.61	3.4	8.9	60	3.3	5.9	10
8. W. Virginia, Babcock State Park	1984	0.81	9.5	3.0	1.1	3.6	14	56	1.4	5.1	11
W. Virginia, Babcock State Park	1985	0.63	6.0	2.3	0.97	2.1	11	58	2.4	3.9	15
9. W. Virginia, Parsons	1982	0.87	11.5	2.9	0.95	3.2	17	74	11	4.2	17
W. Virginia, Parsons	1985	0.63	14	3.5	1.2	2.9	19	97	7.3	5.4	18

* This table is arranged with the first row of data for each site representing a year with a $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratio significantly higher than the next row. Acid fluxes which show a significant increase are underlined.

are (1) a change in the composition of the power plant fuel, (2) a change in normal background SO_4^{2-} and NO_3^- acidities. Even if all the excess Cl^-, i.e., that greater than $Na^+/0.86$, were in the form of HCl, only 1–3% of the total H^+ could be from HCl. Thus the major part of the acidity originates with SO_4^{2-} and NO_3^- and small changes in their background values could overshadow the effects sought.

Central and Appalachian coals of the USA contain from 0.01 to 0.5 wt.% chlorine (Bureau of Mines, 1966). Combustion of these coals could release thousands of tons of Cl^- per year from a single metropolitan power plant (PHS, 1968). A concentration of 49 ppm HCl has been measured in stack gases from a coal containing 0.066% Cl which corresponded to a 60% release of the Cl as HCl (PHS, 1968). The combustion of coal has recently been suggested by Brown et al. (1988) as a possible important source of chlorine for the catalytic decomposition of ozone.

Two recent publications, Lightowlers and Cape (1988) and ten Brink et al. (1988), essentially predict the results in area B. The former reference estimates that 93% of the HCl emissions in the UK are from coal burning. This % is based on detailed estimates of several other sources: waste incineration, chlorinated hydrocarbons, automobile exhausts, glass melting, fuel oil combustion, and steel pickling. Waste incineration is the next most important source of HCl. The references find experimentally and theoretically that plume washout of HCl into rain occurs rapidly near coal-fired power plants and is more rapid and extensive than for S compounds. The USA is culturally and economically similar enough to the UK to expect similar results in the USA, if the chloride contents of the coals are of the same order of magnitude. Lightowlers and Cape (1988) used an average of 0.25% Cl for UK coals which is a median value for eastern USA coals (Bureau of Mines, 1966).

No pattern of low $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratios are found near large cities in the USA or in Southern California, where auto emissions are a problem. In area B over the years 1982–1986, there was no reduction in excess Cl^- coincident with the lowering of the lead content of gasoline: 1.1 g/gal pre-1 July 1985; 0.5 g/gal 1 July 1985 to 1 January 1986; 0.1 g/gal post 1 January 1986. These observations indicate that chlorinated hydrocarbon lead scavengers from automobile exhaust, are not a significant source of Cl^- in rain.

There is positive correlation between excess Cl^-, using Na^+ as the conservative element, and non-sea-salt SO_4^{2-} at only about one-half of the sites in area B. One possible explanation of this is that some Na^+ is washed out of the plume, but Na^+ is not easily extracted by water, at least, from fly ash (Andren et al., 1980). However, a more likely explanation is the very different kinetics of the washout of HCl and S compounds as noted by ten Brink et al. (1988).

Fig. 2 shows the location of coal-burning power plants in the USA relative to the NADP/NTN rain collection sites. Plants with flue gas scrubbers (FGD units) are noted because scrubbers are efficient HCl removers (ten Brink et al., 1988). However, scrubbers can be bypassed, are not on line at all times, and do not in all cases match the coal firing units, unit for unit. Sites with ratios of $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ less than 0.90 are concentrated in the area of highest density of power plants. Generally, the low ratio sites are near one or more coal-burning power plants without scrubbers. This situation is illustrated in Table 2 where the megawatt capacity (MW), distance and direction of the power plants from a selected group of rain collection sites are noted. The six sites in Table 2 were selected because they have consistently low $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratios of 0.90 or less for each year, 1982–1986, as shown in Table 3. These "model" low ratio sites are characterized by having nearby coal-burning power plants in an up-prevailing-wind direction.

Of the total 178 collection sites in Fig. 2, 36 have $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratios less than 0.90. Of these 36 sites, 30 have one or more coal-burning power plants within 50 km. 5 of the remaining 6 have one or more coal-burning power plants within 50–100 km. Only one site, the northern most site in New York (NY) state has no power plant within 100 km. This latter site is at a high elevation (622 m) and in a down wind position from a high emission region.

There are some sites with nearby power plants but with $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratios of 1 or greater. These occur mainly outside area B, Fig. 1, and possibly have just enough extra Na^+ from terrestrial sources to neutralize the HCl effect on the ratio. Several are upwind of the power plant.

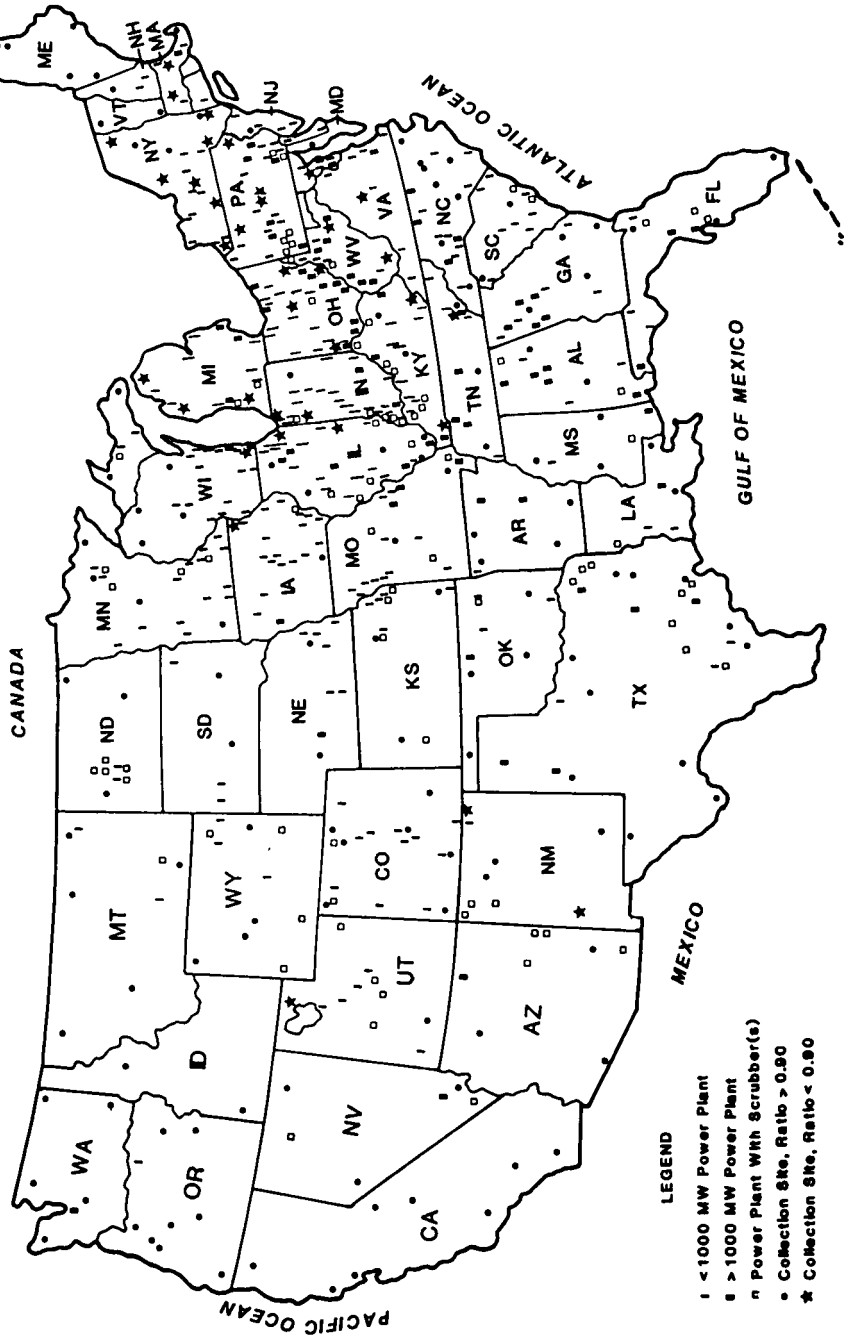

Fig. 2. Location of lignite and coal-burning power plants relative to the collection sites in Fig. 1. Power plant data are from EIA, 1986 and are as of December 1986. Site locations and emission data from Robertson and Wojciehowski, 1986. Plants with scrubbers (FGD) determined from 1988 *ad hoc* communication from *Energy Information Administration*, Washington.

Table 2. *Coal-burning power plants near selected rain collection sites*

Site	Nearest power plants; output (megawatts), distance and direction from collector
Caldwell, OH	1425 MW (23 km SW), 316 MW (64 km SW)
Chautauqua, NY	50 MW (22 km SE), 555 MW (21 km N), 120 MW (69 km SW)
Indiana Dunes National Lakes, IN	589 MW (20 km NE), 480 MW (2 km NE), 485 MW (18 km SW), 490 MW (36 km W)
Parsons, WV	1613 MW (38 km NE), 292 MW (ca. 50 km N), 139 MW (ca. 60 km NW), 1107 MW (ca. 70 km NW)
Walker Branch Watershed, TN	1595 MW (18 km SW), 905 MW (15 km NE), 224 MW (63 km SW)
Wooster, OH	192 MW (14 km NE), 29 MW (46 km SE), 96 MW (51 km NE), 37 MW (57 km ENE)

Table 3. $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ *ratios vs years for sites in Table 2*

	$(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$				
Site	1982	1983	1984	1985	1986
Caldwell, OH	0.53	0.71	0.65	0.60	0.73
Chautauqua, NY	0.63	0.71	0.76	0.76	0.79
Indiana Dunes National Lakes, IN	0.85	0.90	0.74	0.76	0.89
Parsons, WV	0.87	0.77	0.80	0.63	0.69
Walker Branch Watershed, TN	0.85	0.79	0.87	0.67	0.85
Wooster, OH	0.83	0.73	0.72	0.66	0.74

Along the sea coasts in Fig. 1, i.e., Atlantic ocean, Pacific ocean and Gulf of Mexico, the $(Na^+/Cl^-)/(Na^+/Cl^-)_{SW}$ ratio is near 1. Using the data from the 45 sites within 150 km of a sea coast, the following average ratio was obtained, 1.00 ± 0.02 (SD). No evidence for "lost chloride" was indicated.

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