

SEASALT AND POLLUTION INPUTS OVER THE CONTINENTAL UNITED STATES

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Abstract. The average deposition rate of sea salt chloride over the world continents is about $10 \text{ meq m}^{-2} \text{ yr}^{-1}$. Only about $14 \pm 1\%$ of chloride in the pollution-corrected world average river is contributed by seasalt aerosols and the rest from the dissolution of evaporites. The significant increase of the ion concentrations in the Mississippi river from the year 1905 to 1987 was caused by anthropogenic inputs such as fossil fuel burning, common salt consumption, and dissolution of carbonate and silicate rocks by acids derived from acid precipitation.

1. Introduction

A small fraction of seasalt aerosols produced at the ocean-atmosphere interface is transported over the continents and deposited as wet (rain) and dry precipitations. On a global scale, how much of the dissolved major ions in the rivers are contributed from the precipitation of seasalt is quite uncertain. For example, Garrels and Mackenzie (1971) suggested that about 55% of the Cl in the world average river water is of seasalt origin; Conway (1942) 44%; Livingstone (1963) 46%; Holland (1978) $\geq 27\%$; Meybeck (1979) 33%; Meybeck (1983) 72%; and Stallard and Edmond (1981) 18%. The rest should be inputs from the dissolution of evaporites embedded in geological strata and from pollution. Meybeck (1979) estimated the pollution input of Cl^- in the world average river today to be about 30%, but with large uncertainty.

The purpose of this paper is to address the problem of the relative importance of various Cl^- sources again and also to illustrate how the gas emissions from anthropogenic and natural origins affect the chemistry of rain as well as river waters. The continental United States ($\approx 8 \times 10^6 \text{ km}^2$, excluding Alaska) is used as an example.

2. Materials and Methods

Rainwater chemistry in the United States has been systematically monitored at about two hundred stations since 1978. The weekly results are published semi-annually and annually by the Coordination Office of National Atmospheric Deposition Program/National Trend Network (NADP/NTN), National Resource Ecology Laboratory, Colorado State University, Fort Collins. The U.S. Geological Survey also has been publishing annually water resources data (hydrology and water chemistry) for numerous river stations throughout the United States. Based on

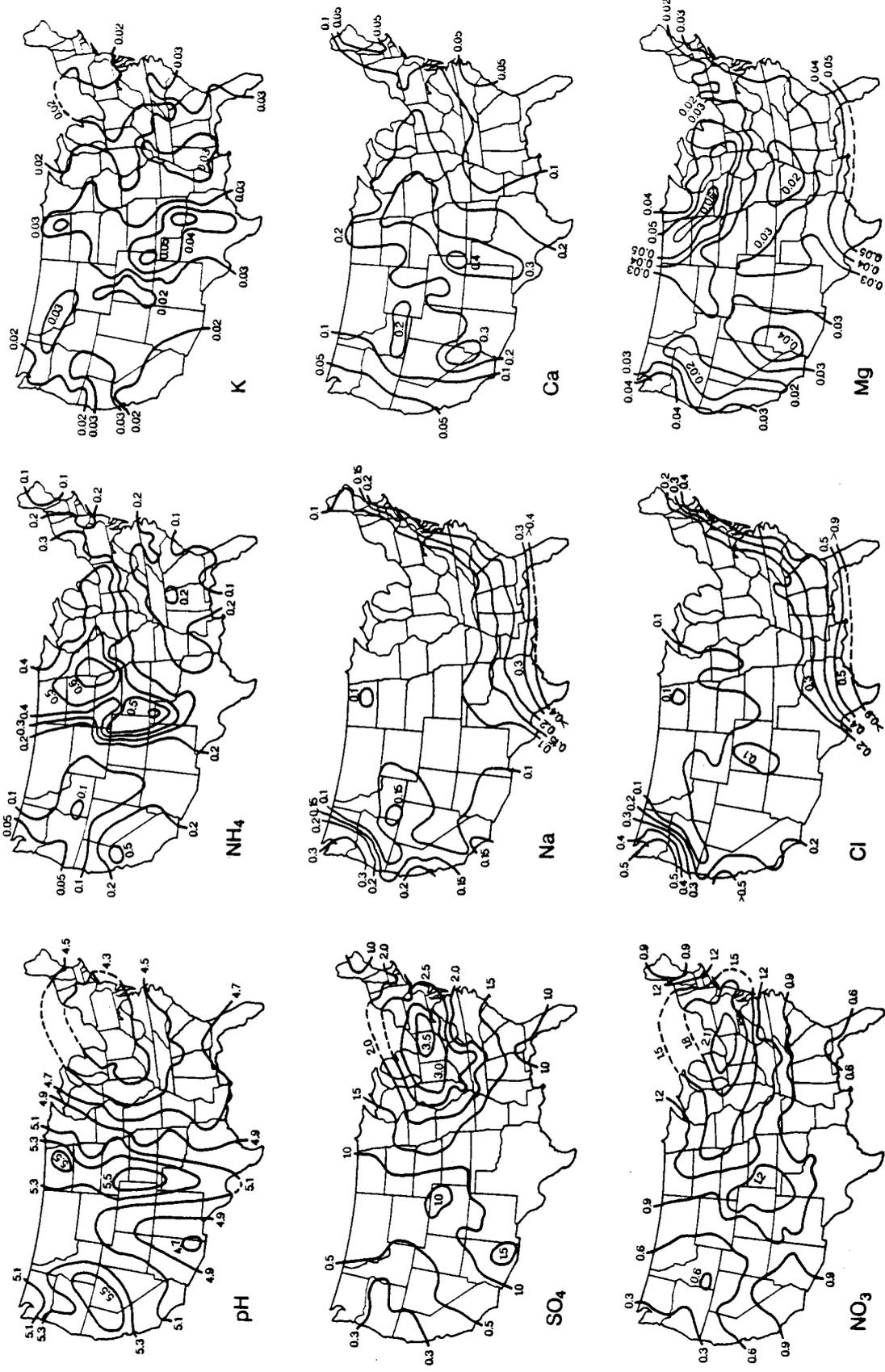


Fig. 1. The isopleth maps for the precipitation weighted mean concentrations of major ions (mg L⁻¹) and pH in rainwaters over the continental United States (modified after figures given in NADP/NTN report-1987).

these data, one can easily estimate the seasalt contribution to the continental U.S. and the Mississippi river basin in particular by simple mass balance calculations. The rainwater data in the period between 1985 to 1989 were studied, but for illustration purpose, the results from 1987 were emphasized.

3. Results and Discussions

SEASALT INPUTS TO THE CONTINENTAL UNITED STATES

Figure 1 summarizes the isopleth maps for the precipitation weighted mean concentrations (mg l^{-1}) of major ions in rainwater, modified after similar figures in the NADP/NTN Annual Data Summary - 1987. The coincidence of the lowest pH and the highest SO_4^{-2} and NO_3^{-} in the northeastern U.S. represents nicely the industrial inputs of SO_2 and NO_x gases from the fossil fuel burning and smelting of sulfide ores. The NH_4^+ concentration is also high in this area, indicating some input of NH_3 gas through fossil fuel burning (Warneck, 1988). However, the concentration of NH_4^+ is highest in the central U.S.A. where the pH is also the highest. The sources of NH_3 gas may be from animal waste from cattle ranches and/or ammonia fertilizer used in the area. The high pH is in part caused by the neutralization of H^+ by NH_3 gas through the overall reaction of $\text{NH}_3 + \text{H}^+ \rightarrow \text{NH}_4^+$.

The rapid decrease in the concentrations of Cl^- , Na^+ and Mg^{+2} from the coasts inland indicates the dominant seasalt inputs for these ions in the coastal areas. However, Mg^{+2} has another maximum over the north-central U.S.A. where Ordovician, Devonian and Silurian dolomitic carbonate rocks are dominant underlying rock types (The National Atlas of U.S.A., 1970). Therefore the major source of Mg^{+2} in the area is the dissolution of dolomite dust particles in rainwater. The highest Ca^{+2} and K^+ rainwater concentrations are again in the central U.S.A., indicating crustal dust and/or fertilizer particle inputs which may also neutralize some H^+ ions in the rainwater through dissolution or ion exchange e.g.,



The precipitation weighted mean concentrations of major ions multiplied by the annual rain precipitation rate give the annual deposition rates of major ions per unit area ($\text{mg m}^{-2} \text{yr}^{-1}$ or $\text{meq m}^{-2} \text{yr}^{-1}$) by rainfall. Figure 2 shows, as an example, the isopleth maps for the annual deposition rates of Cl^- and H^+ during the year 1987. For complete data, one should refer to the NADP/NTN Annual Data Summary - 1987. The dotted line in Figure 2 encloses the Mississippi river drainage basin. From figures like Figure 2, the area weighted mean deposition rates of major ions by rainfall can be estimated for the whole Mississippi river drainage basin. The results are summarized in Table I in units of $\text{meq m}^{-2} \text{yr}^{-1}$. If one assumes that the Cl^- in rainwater is all of seasalt origin, then the percent contribution of seasalt

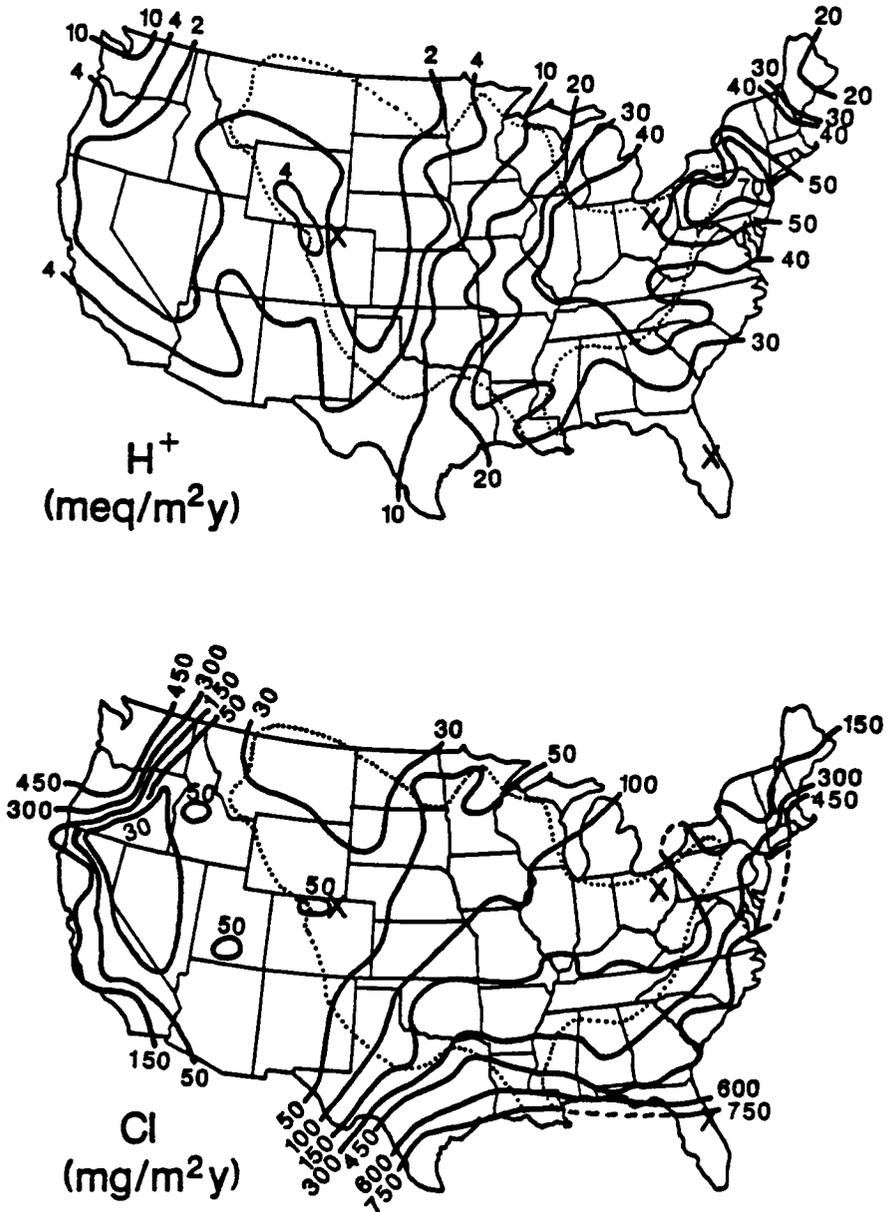


Fig. 2. The isopleth maps for annual deposition rates of H^+ and Cl^- ions per unit area over the continental United States (modified after the NADP/NTN report-1987). The area enclosed by dotted line is the Mississippi river drainage basin. The crosses indicate the rain station locations for the Kennedy Space Center in Florida, Wooster in Ohio, and Pawnee in Colorado as mentioned in the text.

TABLE I

Average deposition rates of ions into the Mississippi river basin from rainwater (1987); river fluxes of ions from the Mississippi river basin (1987, this study, based on USGS water resource data; and 1905, Clarke, 1924); and various pollution inputs. All in unit of meq m⁻² yr⁻¹, except SiO₂ in mmol m⁻² yr⁻¹. Values in parentheses are percent contributions accounted for by seasalt input.

	Mississippi River			Δ (b)-(c)	Fossil fuel input	Common salt input	Carbonate dissolution input	Silicate dissolution input	Rest Δ-Σ input
	Rain 1987 (a)	1987 (b)	1905 (c)						
H ⁺	17.00				78		-58	-20	0
NH ₄ ⁺	12.4								
Na ⁺	3.0 (100)	199 (1.8)	85 (4.1)	114		88		20	6
K ⁺	0.6 (11)	15 (0.5)	13 (0.6)	2					2
Ca ⁺²	7.2 (1.8)	358 (0.04)	300 (0.05)	58			58		0
Mg ⁺²	1.8 (38)	188 (0.4)	130 (0.6)	58			58		0
Cl ⁻	3.5 (100)	143 (2.9)	51 (8.0)	92	4	88			0
SO ₄ ⁻²	25.0 (1.4)	206 (0.2)	132 ^a (0.6)	74	74				0
HCO ₃ ⁻	≈ 0	398	337	61			58		3
NO ₃ ⁻	13.5	13	8	5					5
SiO ₂		20	22						
Σ [±]	42	760	582	178					

Q = 580 km³ yr⁻¹, A = 3.27 × 10⁶ km², Q/A = 177 L m⁻² yr⁻¹.

^a sulfate is obtained by charge balance.

Σ[±] = total cation or anion.

in rainwater for each major ion can be estimated (Table I, parentheses). The average seasalt contributions of Ca^{+2} and SO_4^{-2} in rainwater over the Mississippi river basin are less than 2%, whereas Na^+ , Mg^{+2} , K^+ are, respectively, about 100, 38, and 11%. The output fluxes of major ions from the Mississippi river to the ocean during the year 1987 and 1905 are also summarized in Table I in units of $\text{meq m}^{-2} \text{ yr}^{-1}$. These fluxes are calculated from the discharge weighted mean ion concentration in the Mississippi river (data from USGS Water Resource Data for Louisiana-1987, station near St. Francisville; and Clarke, 1924, for the 1905 data) multiplied by the long term average discharge rate of the river ($Q=580 \text{ km}^3 \text{ yr}^{-1}$) and divided by the river's drainage area ($A=3.27 \times 10^5 \text{ km}^2$) (Milliman and Meade, 1983).

How much of seasalt aerosol is deposited on the continent as dry precipitation is quite uncertain. For example, Lindberg *et al.* (1990) found that the dry deposition washed off from forest canopy (so-called net throughfall) for Cl^- is about 17 and 135% of the wet deposition rate, respectively, at Oak Ridge, Tennessee and Göttingen, Germany. The net throughfall for Cl^- can be as high as 2 to 3.5 times the wet deposition rate for forest canopy near coastal areas in England (White and Turner, 1971). One may generalize that the dry/wet Cl^- deposition ratio is low in the inland regions and high in coastal regions. If one assumes that the inland Oak Ridge station is representative of the whole Mississippi river basin with regard to the relative contribution of wet and dry Cl^- deposition, the total seasalt Cl^- contribution to the Mississippi river is estimated to be only 3% of the river Cl^- flux in 1987 and 8% in 1905 (Table I). Even assuming the dry Cl^- deposition

TABLE II

The average river output fluxes of ions from Amazon basin, North America and the world continents (all in $\text{meq m}^{-2} \text{ yr}^{-1}$ except SiO_2 in $\text{mmol m}^{-2} \text{ yr}^{-1}$) and percent contribution of seasalt for each chemical species (in parentheses).

	Amazon basin (Gibbs, 1972)	North America (Meybeck, 1979)	World average (Meybeck, 1979)
Na^+	121 (9)	101 (8)	95 (9)
K^+	23 (1)	14 (1)	14 (1)
Ca^{+2}	292 (0.2)	360 (0.1)	282 (0.1)
Mg^{+2}	74 (3)	145 (1)	116 (1)
Cl^-	99 (13)	71 (14)	68 (15)
SO_4^{-2}	56 (2)	111 (1)	73 (1)
HCO_3^-	332	420	360
NO_3^-	3		
SiO_2	168	43	73
Σ^+	510	620	507
Σ^-	490	602	501
Q ($10^3 \text{ km}^3 \text{ yr}^{-1}$)	5.51	5.53	37.4
A (10^6 km^2)	6.13	15.4	88.6
Q/A ($\text{L m}^{-2} \text{ yr}^{-1}$)	899	359	422

rate equal to the wet Cl^- deposition rate, the Cl^- contribution is only 5% in 1987 and 14% in 1905.

The area weighted mean Cl^- deposition rate from the rain over the continental United States is estimated to be about $5.0 \text{ meq m}^{-2} \text{ yr}^{-1}$ in 1987 (using Figure 2; this value ranges from 4.8 to $5.8 \text{ meq m}^{-2} \text{ yr}^{-1}$ with an average of $5.3 \text{ meq m}^{-2} \text{ yr}^{-1}$ in the period between 1985 to 1989). If one assumes the dry Cl^- deposition rate is roughly equal to the wet Cl^- deposition rate on a continental scale (average out high dry/wet ratio in coastal area and low dry/wet ratio inland i.e., the total Cl^- deposition rate of $10 \text{ meq m}^{-2} \text{ yr}^{-1}$), the total seasalt Cl^- contribution to the average pollution-corrected North America River Water (Maybeck, 1979) would still be only about 14% (Table II).

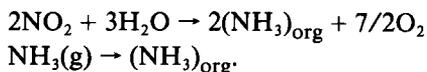
Stallard and Edmond (1981) estimated the mean seasalt Cl^- concentration ($\overline{\text{Cl}}$) in the Amazon river (supposedly less polluted) as a whole to be about $9.2 \mu\text{mole L}^{-1}$ at the time of peak discharge (wet season) in the period of 1976 to 1977. This seasalt Cl^- is supposed to include both wet and dry depositions. The Amazon river drainage area (A) above Obidos is about $5 \times 10^6 \text{ km}^2$ (Oltman *et al.*, 1963, 1968) and the discharge rate (Q) at Obidos was $230\,000 \text{ m}^3 \text{ s}^{-1}$ (i.e., $725 \times 10^{13} \text{ L yr}^{-1}$) at the peak discharge during the same period (Stallard and Edmond, 1981). Therefore, the Q/A ratio is about $1440 \text{ L m}^{-2} \text{ yr}^{-1}$. If this ratio also applies to the whole Amazon river basin (which has a drainage area of $6.13 \times 10^6 \text{ km}^2$), the mean seasalt Cl^- input (both dry and wet) per unit area of the Amazon river basin should be about $13 \text{ meq m}^{-2} \text{ yr}^{-1}$ ($= \overline{\text{Cl}} \times Q/A$) at peak discharge, which is similar to the value estimated for the continental United States in this study. Even the deposition rate of seasalt Cl^- during the dry season is assumed to be equal to that of wet season, the cyclic seasalt Cl^- would account for only 13% of the Cl^- flux in the Amazon river (Table II; the discharge weighted mean concentrations of ions in the Amazon river are from Gibbs, 1972). Stallard and Edmond (1981) gave 18% for the Amazon river above Obidos at peak river discharge (wet season).

Using the average seasalt Cl^- deposition rate of $10 \text{ meq m}^{-3} \text{ yr}^{-1}$ for the world continents and the pollution-corrected World Average River Water composition given by Meybeck (1979), seasalt Cl^- accounts for only 15% of Cl^- river flux worldwide (Table II). Also the mean residence times of Cl^- and Na^+ in the ocean would be about 140×10^6 and $83 \times 10^6 \text{ yr}$, respectively, after correcting for the cyclic seasalt components. The overestimation of cyclic seasalt in the world average river water by earlier works was probably in part biased by many smaller coastal river basins which contain large seasalt inputs as compared to the continental scale river basins, a suggestion made also by Berner and Berner (1987).

POLLUTION INPUTS TO THE MISSISSIPPI RIVER

The increases in the ion concentrations or fluxes for the Mississippi river from the year 1905 to 1987 (Table I) can be mostly explained as being due to anthropogenic inputs. For example, the increment of $74 \text{ meq SO}_4^{-2} \text{ m}^{-2} \text{ yr}^{-1}$ is most probably due to input from fossil burning ($\text{SO}_2 + \text{H}_2\text{O} \rightarrow 2\text{H}^+ + \text{SO}_4^{-2}$; ignoring many

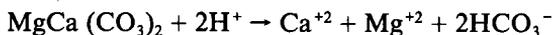
intermediate steps). The coal and crude oil consumptions for the United States (\approx production + net import) were, respectively, 0.83×10^{15} g yr⁻¹ and 1.1×10^{15} g yr⁻¹ in 1987 and negligible in 1905 as compared to 1987 (Statistical Data of the United States, 1941, 1989). If the S contents of coal and crude oil are on the order of 1 and 0.1% respectively, the burning of fossil fuel would release on the order of 100 meq SO₄⁻² m⁻² yr⁻¹ over the continental United States, in good agreement with the observed 74 meq SO₄⁻² m⁻² yr⁻¹ in the Mississippi river basin, considering some atmospheric export of anthropogenic S to the Atlantic Ocean and Canada (Bischoff *et al.*, 1984). Introduction of 74 meq SO₄⁻² m⁻² yr⁻¹ through fossil fuel burning should also result in the release of about 4 meq Cl⁻ m⁻² yr⁻¹ (Lightowers and Cape, 1988) and 78 meq H⁺ m⁻² yr⁻¹ (= 74 + 4). Most of NO_x and NH₃ gases released by fossil fuel burning did not show up in the Mississippi river as dissolved NO₃⁻ (Table I); thus, probably end up as NH₃ in living organic matter e.g.,



Interestingly, if the dry deposition is twice as much as the wet deposition for SO₄⁻², H⁺ and NH₄⁺, the total atmospheric deposition of SO₄⁻², H⁺ and NH₄⁺ (NH₄⁺ → NH₃_{org} + H⁺) would explain the anthropogenic inputs of SO₄⁻² and H⁺ in the Mississippi river basin.

The increments of 88 meq m⁻² yr⁻¹ for Cl⁻ and associated Na⁺ are probably due to human input from common salt (NaCl) consumption. The consumption rate (\approx production rate) of common salt in the United States was about 3.3×10^{12} g yr⁻¹ and 36×10^{12} g yr⁻¹ during the year 1905 and 1987, respectively (Statistical Abstracts of the United States, 1941, 1989). Therefore, from 1905 to 1987, the increment of common salt input in the continental United States was on average about 72 meq NaCl m⁻² yr⁻¹, which is again in close agreement with the observed value (88 meq m⁻² yr⁻¹) for the Mississippi river basin. The anthropogenic inputs of Cl⁻ in the Mississippi river account for 64% of the total Cl⁻ in 1987 as against the 30% for the world average river (Meybeck, 1978).

The increments of 58 meq m⁻² yr⁻¹ for Ca⁺², Mg⁺² and associated HCO₃⁻ can be easily explained by the dissolution of dolomite through the reaction:



where H⁺ comes from fossil fuel burning. The rest of H⁺ (20 meq m⁻² yr⁻¹) can replace Na⁺ in silicate mineral particles by ion exchange and dissolution processes. The small leftover HCO₃⁻, Na⁺, K⁺, and NO₃⁻ in Table I (column 'Rest') can be attributed to a slight increase in natural chemical weathering rate and fertilizer inputs or simply data noise.

CORRELATION AMONG THE ION CONCENTRATIONS IN RAINWATERS

Four stations from the NADP/NTN Annual Data Summary were selected to compare their rainwater chemistry: American Samoa in the Pacific ocean (least polluted),

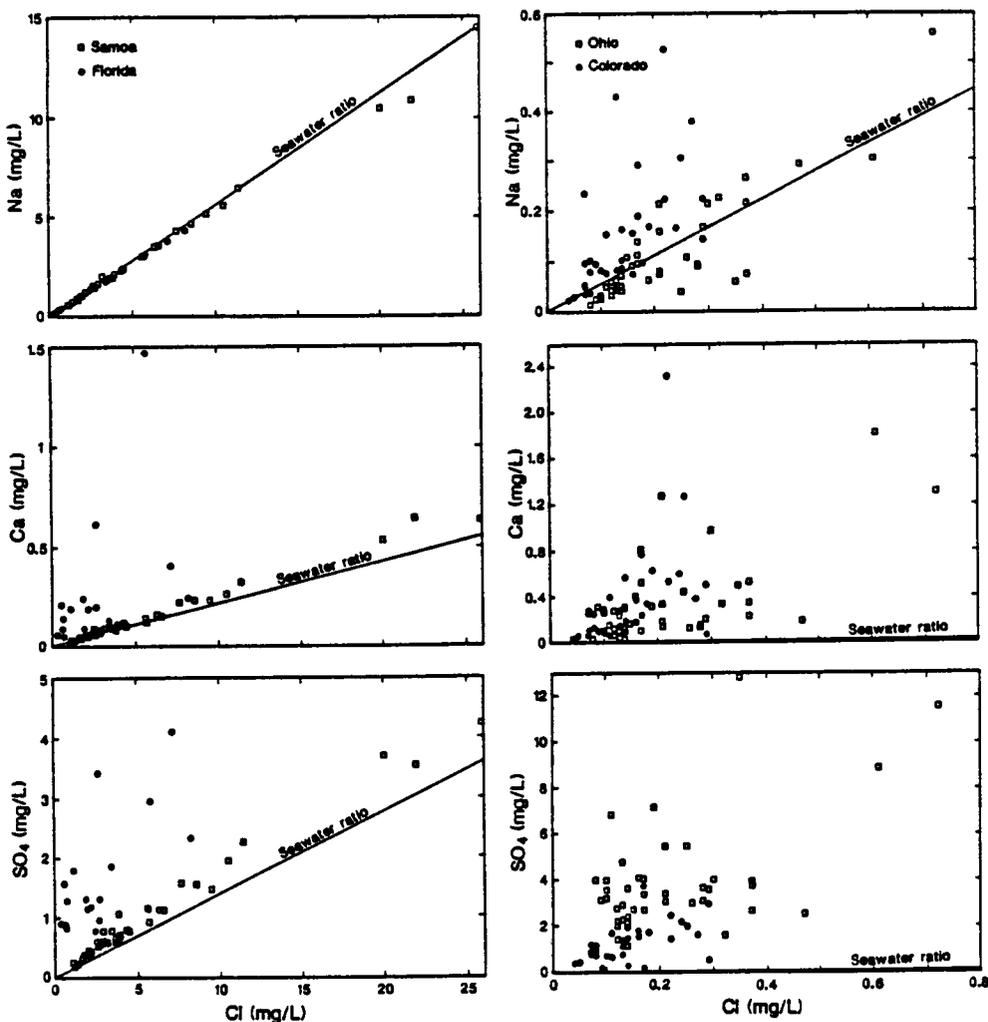


Fig. 3. The plots of the weekly concentration data of chloride versus those of Na^+ , Ca^{2+} , and SO_4^{-2} from American Samoa and Florida at Kennedy Space Center (left side); and Ohio at Wooster and Colorado at Pawnee (right side). The solid lines indicate seawater ratios. Data are from NADP/NTN 1987 report except American Samoa, 1989.

Florida station at Kennedy Space center ((high seasalt), Ohio station at Wooster (highest SO_4^{-2} , NO_3^- , and H^+ concentrations) and Colorado station at Pawnee (highest NH_4^+ concentration). The locations of the last three stations are indicated as crosses in Figure 2.

The precipitation weighted mean compositions of rainwater from these four stations are summarized in Table III along with their seasalt corrected compositions. Some of their weekly rainwater data are plotted in Figure 3. Figure 4 also summarizes

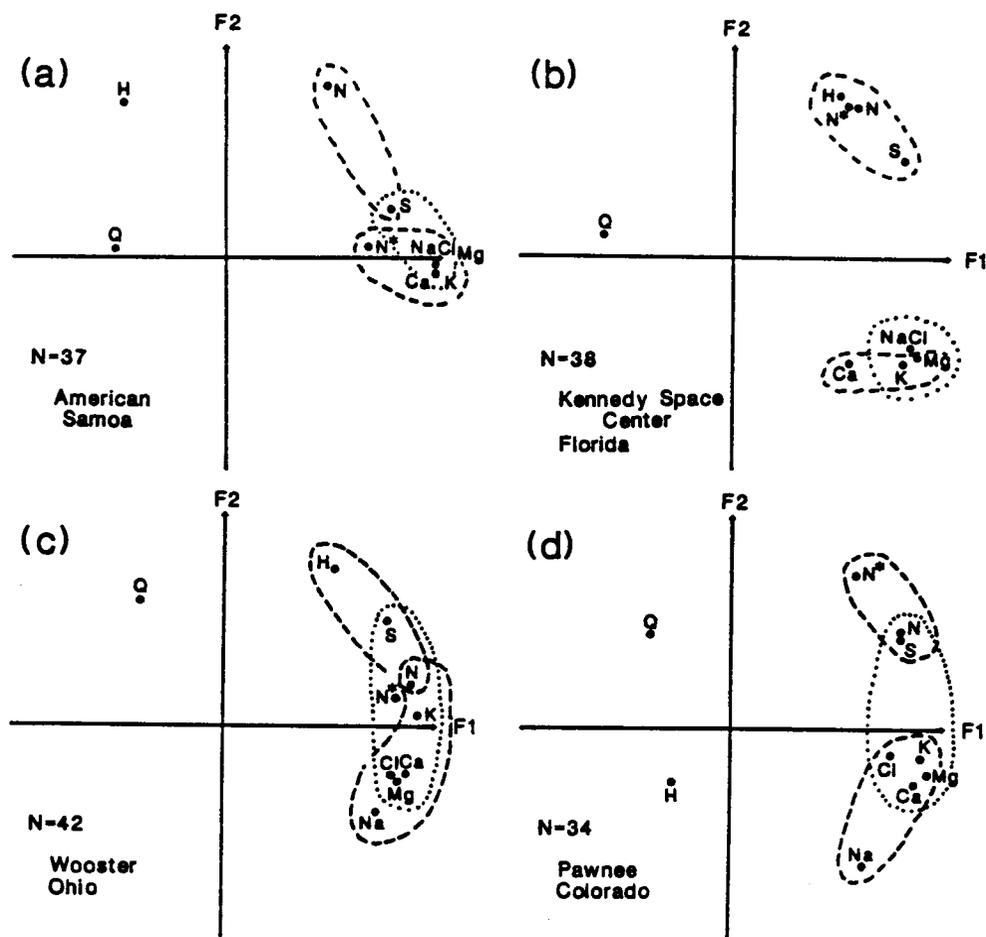


Fig. 4. The plots of the principal component factor 1 versus factor 2 loadings of various ions at four different stations, using weekly rainwater chemistry data (NADP/NTN 1987 report except American Samoa, 1989) where N, N*, S, H, and Q represent respectively NO_3^- , NH_4^+ , SO_4^{2-} , H^+ and precipitation rate. Elements within a dotted loop have correlation coefficients of greater than 0.5 between any pair of them. The $N=xx$ represents the sample numbers used in the factor analysis.

the results of principal component factor analyses (using the weekly rainwater composition data) to show the interelemental relationship in the rainwater. Elements within a dotted loop of Figure 4 are highly related (correlation coefficients of greater than 0.5 between any pair of them). The Q, N, N*, H, and S in Figure 4 represent, respectively, precipitation rate of rainwater, the concentrations of NO_3^- , NH_4^+ , H^+ , and seasalt-corrected (or called non-seasalt) SO_4^{2-} .

As one would have expected, the American Samoa rains contain mostly seasalt with only small amounts of non-seasalt components such as H^+ , SO_4^{2-} , NH_4^+ , NO_3^- , and Ca^{+2} (Table III). Their Na/Cl ratios are similar to the seawater ratio, whereas their Ca/Cl and SO_4/Cl ratios are slightly higher than the seawater ratios (Figure 3).

TABLE III

Precipitation weighted mean composition of rainwater from selected sites (NADP/NTN, 1987; except American Samoa, 1989) and their seasalt corrected compositions ($\mu\text{eq L}^{-1}$)

	Seasalt equivalent ratio	American Samoa	Florida (Kennedy Space Center)	Ohio (Wooster)	Colorado (Pawnee)
H ⁺		3.0	17.8	60.2	3.9
NH ₄ ⁺		1.1	4.4	27.7	36.1
Na ⁺	0.857	72.9	32.0	2.3	4.1
K ⁺	0.018	1.6	0.9	0.5	1.1
Ca ⁺²	0.038	4.0	4.5	10.4	13.4
Mg ⁺²	0.195	16.4	7.6	3.0	2.7
Cl ⁻	1	86.0	38.4	3.9	3.1
SO ₄ ⁻²	0.103	11.9	21.2	75.4	24.8
NO ₃ ⁻		0.6	9.7	33.4	25.0
Σ ⁺	1.108	99.0	67.2	104.1	61.3
Σ ⁻	1.103	97.8	69.2	112.7	52.9
H ⁺		3.0	17.8	60.2	3.9
NH ₄ ⁺		1.1	4.4	27.7	36.1
Na ⁺		-0.8	-0.8	- 1.0	1.5
K ⁺		0.1	0.2	0.5	1.0
Ca ⁺²		0.7	3.0	10.2	13.4
Mg ⁺²		-0.4	0.1	2.2	2.1
Cl ⁻		0	0	0	0
SO ₄ ⁻²		3.0	17.3	75.0	24.4
NO ₃ ⁻		0.6	9.7	33.4	25.0
Σ ⁺		3.7	24.7	99.8	58.0
Σ ⁻		3.6	27.0	108.4	49.4

The source of small non-seasalt Ca⁺² is probably the dissolution of carbonate dust (from coral). The most probable source for the non-seasalt NH₄⁺ and SO₄⁻² is the surface ocean. As shown by Quinn *et al.* (1990) and Kiene and Bates (1990), the surface oceans are important sources for NH₃ and dimethylsulfide (DMS) gases in the oceanic atmosphere, and DMS is eventually oxidized to SO₄⁻². The close correlation of non-seasalt SO₄⁻² and NH₄⁺ with major seasalt ions (Figure 4a) is consistent with this explanation. On the other hand, the weak correlation between non-seasalt SO₄⁻² and NO₃⁻ (=0.54) suggest that the sources of NO₃⁻ may not entirely be related to that of SO₄⁻² and NH₄⁺. Some NO₃⁻ could be originated from the oxidation of organic compound in the air, lightning and stratosphere (Savoie and Prospero, 1989a, b; Prospero and Savoie, 1989; Levy II and Moxim, 1989). Dayan and Nelson (1988) also suggest that some natural non-seasalt SO₄⁻² and H⁺ (from organic acids such as formic and acetic acids) may be transported from Australia and New Guinea. One cannot, however, totally rule out that small fractions of NO₃⁻ and SO₄⁻² are anthropogenic (Levy II and Moxim, 1989). In contrast, most

of NO_3^- and SO_4^{2-} in the Bermuda rainwater are of anthropogenic inputs from North America and European continents (Galloway *et al.*, 1989; Levy II and Moxim, 1989).

Two thirds of dissolved ions in the Florida rains are seasalts and the rest non-seasalt H^+ , NH_4^+ , SO_4^{2-} , NO_3^- , and Ca^{+2} (Table III). The Na/Cl ratios are close to the seawater ratio, but Ca/Cl and SO_4/Cl ratios are much higher than the seawater ratios (Figure 3). The high correlation among the concentrations of non-seasalt SO_4^{2-} , H^+ , NH_4^+ , and NO_3^- (Figure 4b) strongly suggests that the source of these constituents is fossil fuel burning (Warneck, 1988). The source of non-seasalt Ca^{+2} is again dissolution of carbonate dust.

In the Ohio rains, the seasalts account for only 4% of the total dissolved ions. Ions other than Cl^- and Na^+ are mostly inputs from fossil fuel burning and dissolution of carbonate and silicate dust (Table III; Figure 3-right). The Na/Cl ratios in Ohio rainwaters vary widely, and more than one half of the data points are less than the seawater ratio (Figure 3-right). The implication is that some of the Cl^- has a non-seasalt origin, and most likely come from fossil fuel burning (Lightowers and Cape, 1988; Wagner and Steele, 1989). The close correlation of Cl^- with non-seasalt SO_4^{2-} , NO_3^- , NH_4^+ , Ca^{+2} , Mg^{+2} , and K^+ (Figure 4c) is also consistent with the above suggestion and furthermore points to dissolution of carbonate and silicate dust by anthropogenic H^+ to release Ca^{+2} , Mg^{+2} , and K^+ into the rainwater. The close correlation among H^+ , SO_4^{2-} and NO_3^- (Figure 4c) indicates that the acidity of Ohio rainwater is mainly controlled by SO_2 and NO_x inputs.

The interelemental relationship in the Colorado rain (Figure 4d) are similar to those for the Ohio rain (Figure 4c), except that Colorado rain has extra NH_3 gas input from cattle ranches and ammonia fertilizers. The NH_3 gas has neutralized some of H^+ , thus the acidity of Colorado rain no longer correlates with the NO_3^- and SO_4^{2-} concentrations (Figure 4d).

4. Conclusions

(1) On a continental-wide scale, the average deposition rate of seasalt chloride is on the order of $10 \text{ meq m}^{-2} \text{ yr}^{-1}$. Contributions from wet and dry depositions are probably roughly equal.

(2) Only about $14 \pm 1\%$ of the chloride in the pollution-corrected North America average river, Amazon river and world average river is contributed by seasalt aerosols. This value is lower than earlier estimates. Most of the chloride in large rivers originated from the dissolution of evaporites.

(3) Excluding the atmospheric seasalt and pollution inputs, the mean residence times of Cl^- and Na^+ in the ocean are about 140×10^6 and 83×10^6 yr, respectively.

(4) The significant increase of major ion concentrations in the Mississippi river from the year 1905 to 1987 can be semi-quantitatively explained by anthropogenic inputs such as common salt consumption, fossil fuel burning, dissolution of carbonate and silicate rocks by acids derived from fossil fuel burning.

(5) Over the least polluted oceanic islands such as American Samoa, rainwater chemistry is mainly controlled by seasalt aerosols, and NH_3 and DMS gas inputs from the surface ocean.

(6) Rainwater chemistry in the inland continental United States is currently mainly controlled by gas emission inputs from fossil fuel burning (SO_2 , NO_x , NH_3 , HCl), cattle ranches and ammonia fertilizers (NH_3); dissolution of carbonate and silicate dust by anthropogenic acids; and ion exchange with silicate dust. Seasalt inputs are minor.

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