Composition of Atmospheric Precipitation

II. Sulfur, chloride, iodine compounds. Bibliography

By ERIK ERIKSSON
Institute of Pedology and Institute of General and Inorganic Chemistry,
Royal Agricultural College of Sweden, Uppsala

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C. Sulfur in rainwater

Unlike the nitrogen most sulfur determinations in rainwater have been carried out comparatively recently. The greater part of the investigations were carried out in the U.S.A. where an extensive survey of the contribution of atmospheric sulfur to the soil has been made for agricultural purposes.

1. Methods and analysis

The analytical procedure has mostly involved a precipitation of the sulfur compounds as $BaSO_4$. If properly done this is a satisfactory procedure. A division between sulfate sulfur and other forms of sulfur has rarely been done. If other forms of inorganic sulfur are present in rainwater they are generally rapidly oxidized by dissolved oxygen to sulfate.

The sampling of rainwater for sulfur analysis has mostly been done in metal containers which, as ALWAY et al. (8) pointed out in 1937 may give too large figures for the sulfur in rain. Most metals adsorb SO_2 directly from the air which is then oxidized to SO_3 and washed down into the containers with the rainwater as H_2SO_4. In areas of a high atmospheric pollution the contribution by this adsorbtion can amount to six times the sulfur actually present in rainwater.

It is obvious that under such circumstances most of the figures on the amount of sulfur in rainwater are too high. An absorption of SO_2 by the soil itself and even by the plants can, however, also occur. (THOMAS 283, 284.) Therefore the rainwater analysis may represent the total sulfur income by soil which from agricultural point of view is of most interest. But in the discussion on the origin of sulfur in precipitation they are of but little use.

The yearly averages from different states within the U.S.A. are shown in table 6 and results from other parts of the world are in table 7.

From table 6 it will be seen that the average yearly amounts of sulfur vary very much even within the same state due to differences in atmospheric pollution. There are also significant differences between different states (or probably better, between different investigations) which at least partly may be due to different sampling techniques (different sampling gauges). There are some striking differences between different states, compare e. g. Alabama, with Indiana. Both are fairly recent.

The U.S.A. investigations cover the eastern central area of the country.

The European figures are generally lower than those from the U.S.A. Some of the Russian figures are low and may therefore correspond to fairly uncontaminated air in continental areas. The New Zealand figures

Table 6. Sulfur in precipitation in the United States and Canada.

Ref.	Place	Period	Precipitation mm	mg S/l	kg S/ha
	Alabama				
294	Auburn	1940—1943	—	—	6.0
294	Birmingham	1940—1943	—	—	34.2
294	Block Belt Subst. Marion Jet	1940—1943	—	—	6.3
294	Exp. field Alexandria	1940—1943	—	—	6.3
294	» » Aliceville	1940—1943	—	—	5.6
294	» » Monroeville	1940—1943	—	—	4.8
294	» » Prattville	1940—1943	—	—	5.6
294	Gulf Coast Subst. Fairhope	1940—1943	—	—	6.5
294	Kinston	1940—1943	—	—	3.2
294	Sand Mountain Subst. Crossville	1940—1943	—	—	6.8
294	Wiregrass Subst. Headland	1940—1943	—	—	4.6
	Georgia				
294	Experiment	1940—1943	—	—	7.8
	Illinois				
95	Chicago	1921/22—1922/23	—	—	234
95	Liberty	1921/22—1922/23	—	—	61.6
279	Urbana	1913—1917	—	—	50.5
	Indiana				
26	Branchville	1946/47	—	—	34.0
26	Evansville	1946/47	—	—	32.5
26	Gary	1946/47	—	—	142
26	Indianopolis	1946/47	—	—	36.5
26	Lafayette	1946/47	—	—	24.3
26	Larwill	1946/47	—	—	26.0
26	Monterey	1946/47	—	—	30.6
26	Montgomery	1946/47	—	—	31.0
26	Orland	1946/47	—	—	28.9
26	Rising Sun	1946/47	—	—	33.2
	Iowa				
100	Ames	1921	—	—	16.7
15, 117, 161, 235, 246, 269, 290, 306, 313.	Mount Vernon (10 years)	1913/14—1931/32	—	—	22.7
	Kentucky				
153	Greenville	1921/22—1922/23	—	—	40.3
153	Lexington, center	1921/22—1922/23	—	—	46.1
153	» , 15 miles from center	1921/22—1922/23	—	—	26.6
153	Lincoln Inst. Shellbyville	1921/22—1922/23	—	—	19.1
153	Mayfield	1921/22—1922/23	—	—	29.0
153	Paducale	1921/22—1922/23	—	—	38.0
153	Russelville	1921/22—1922/23	—	—	40.3
	Minnesota				
8	Minneapolis, Weather Bureau	1936—1937	—	—	170
8	» , University Farm	1936—1937	—	—	31.1
8	Page	1936—1937	—	—	8.2
8	Bemidji	1936—1937	—	—	5.0
	Mississippi				
294	State College	1940—1943	—	—	5.8

Ref.	Place	Period	Precipitation mm	mg S/l	kg S/ha
	New York				
310	Alfred	1923/24—1924/25	—	—	55.6
310	Brockport	1923/24—1924/25	—	—	86.7
75	Geneva	1919—1928	—	—	46.2
308, 309, 310.	Ithaca	1915/16—1925/26	—	—	38.3
	Oklahoma				
132	Goodwell	1933—1934	—	—	6.5
132	Guthrie	1931—1939	—	—	9.0
132	Heavener	1934	—	—	19.0
132	Lone Grove	1934, 1936	—	—	10.8
132	Stillwater	1927—1942	—	—	9.9
	Tennessee				
195	Columbia	1919—1921	—	—	29.5
195	Copperhill,	1919—1921	—	—	260
195	» , 6 miles from center	1919—1921	—	—	80.9
195	Crossville	1919—1921	—	—	14.2
195	Jackson	1919—1921	—	—	66.2
294	Jackson	1940—1943	—	—	14.8
195	Knoxville, in town	1919—1920	—	—	103
195	» , University Farm	1917—1920	—	—	48.0
195	» , 7 miles from center	1917—1920	—	—	20.7
195	London	1919—1921	—	—	21.6
195	McGee	1919—1921	—	—	27.1
	Texas				
114	Angleton	1924—1928	—	—	10.6.
114	Balhorhea	1924—1928	—	—	4.6
114	Beaumont	1924—1928	—	—	17.2
114	Beeville	1924—1928	—	—	7.4
114	Chillicothe	1924—1928	—	—	11.2
114	College Sta. (Main)	1924—1928	—	—	14.2
114	Denton	1924—1928	—	—	13.2
114	Lubbock	1924—1928	—	—	13.4
114	Nacogdoches	1924—1928	—	—	13.4
114	San Antonio	1924—1928	—	—	9.2
114	Spur	1924—1928	—	—	9.0
114	Temple	1924—1928	—	—	10.0
114	Troup	1924—1928	—	—	10.2
114	Weslaco	1924—1928	—	—	17.4
	Virginia				
99	Blacksburg	1923—1928	—	—	18.9
	Wisconsin, Canada				
135	Madison	1910	—	—	7.8

are fairly representative of conditions in uncontaminated air in coastal places.

The yearly variation is as a rule greater in areas of a high degree of atmospheric pollution than in areas of a low.

The monthly variation is interesting from the point of view of the origin of sulfur in rainwater. In densely populated areas the winter months are highest in sulfur due to increased coal consumption during this season.

In the results from several years of investigation there are sometimes trends, the values increase during the period, indicating an in-

Table 7. Sulfur in precipitation in Europe and other countries except America.

Ref.	Place	Period	Precipitation mm	mg S/l	kg S/ha
	Europe				
27, 28	*France*: Paris	1931/32	(600)	16.0	(96)
27, 28	Grignon	1934/35	(600)	2.5	(15)
188	*Germany*: Müncheberg	1933/34	405	3.3	13.4
213	*Great Britain*: Rothamsted	1881/82—1886/87	760	1.0	7.9
81	Garforth	1907/08	720	1.4	10.2
312	*Russia*: Leningrad	1909/10	646	4.9	31.5
312	» outskirt of town	1909/10	669	4.8	31.7
	Pawlosk	1909/10	522	1.4	7.3
	Sapolje, Kreise Luga	1909/10	486	.9	4.3
	Smolensk	1909/10	660	.4	2.7
	Jekaterineslaw	1909/10	339	6.5	22.1
	Schotilowska, Tule distr.	1909/10	476	.8	3.7
	Borowoje, Ssarnara	1909/10	541	.4	2.3
	New Zealand				
125	Lincoln, Canterbury	1884—1887	750	.9	6.8
	Asia				
216	Tokio, Japan	1938	—	1.9	—

creasing degree of atmospheric pollution. This can be observed in the figures from Ithaca, N.Y. (310) where the yearly amount increases regularly from 1919—20 to 1925—26. In other places, e.g. in Stillwater, Oklahoma (132) no regular trend can be observed but rather periodic fluctuations. The variation in the yearly amounts in Ithaca and Stillwater is shown in fig. 7.

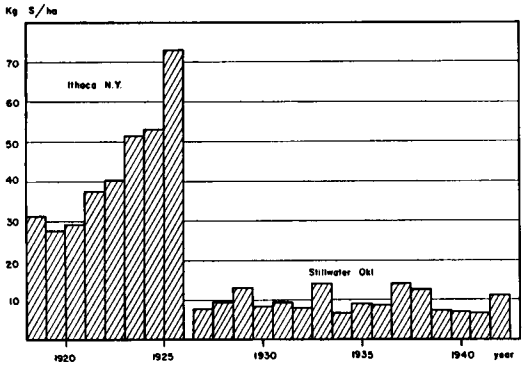

Fig. 7. The yearly amounts of sulfur in precipitation in Ithaca, N.Y. and Stillwater, Okl.

2. Origin of combined sulfur in the rainwater

As mentioned before sulfur present as sulfate has usually been estimated in the analytical procedure. In one instance (81) sulfide, sulfite and organic sulfur have also been determined besides sulfate sulfur. But then no division between sulfide, sulfite and organic sulfur was made.

There is probably very little of sulfide and sulfite present in rainwater, as H_2S is rather volatile and probably exists only as a gas in the air. Sulfite is rapidly oxidized to sulfate by dissolved oxygen in the rainwater but may be present in the air in appreciable concentrations.

There are several probable sources of sulfur in the atmosphere. One of these is coal which on combustion releases considerable amounts of sulfur. In industrial areas it has been noted that the yearly amounts of sulfur brought down by precipitation is very high. There are a number of recent investigations carried out to asses the degree of atmospheric pollution in industrial centres; but these will not be referred to in this paper. One of the earlier

investigations, however, of great agricultural interest was carried out in Leeds and its vicinity around 1910 by CROWTHER & RUSTON (81) (see also CROWTHER & STEWART 82). The results show very clearly that sulfur in precipitation is a very sensitive indicator of atmospheric pollution caused by industries. Even at 5 km distance from the centre of Leeds 170 kg S per hectare and year was brought down by precipitation and at 11 km distance it varied between 57 and 108 kg.

Apart from coal, sulfide ores used in industry certainly contribute to the sulfur content of the air.

Another source is organic matter which, on decay may give off hydrogen sulfide to the atmosphere which is oxidized to SO_3 and dissolved in the rainwater. The rate of delivery of hydrogen sulfide in this way will depend on the season. Hydrogen sulfide, formed by the reduction of sulfate in the sea is another source of atmospheric sulfur as suggested by CONWAY (78).

A great source for atmospheric sulfur is, no doubt, the sea. The sulfate is brought into the air by spray and precipitated by rain water. In coastal areas it is therefore interesting to study this contribution from the sea.

It is fairly well accepted that the main source for chloride in rainwater is the sea, and the only way it can be derived from there seems to be by spray. Comparing the ratio Cl/S in rainwater and in sea water in coastal districts, GRAY (125) in New Zealand observed, however, that this ratio was smaller for rainwater than for sea water. As the place where he collected the rainwater was situated on a small peninsula far from industries it could be expected that the ratio would be the same, as long as the proportion of chloride and sulfate is the same in the spray from the sea as in the sea water. In table 8 some values of the Cl/S ratio from different places are tabulated. It is seen that the New Zealand values come nearest to the expected values. There is also a seasonal variation, the winter and spring values being highest. This can be due to an accelerated liberation of H_2S during the summer and the autumn, not only from the soil but also from the sea.

It is remarkable that even the highest values of the Cl/S ratio in rainwater are only half as great as in sea water. Unless the washing out

Table 8. The ratio Cl/S in precipitation in some places.

Ref.	Place	Cl/S
125	Lincoln, Canterbury New Zealand	
	Sept. to Nov.	11.05
	Dec. to Febr.	9.86
	March to May	9.00
	June to Aug.	10.89
281	Sugashima, Japan at the see	5.85
281	Nagoya, » 4000 m from the sea	6.31
281	Matsumato, » 100 km » » »	.92
170	Holland 440 m from the sea	8.70
170	» 2280 » » » »	5.70
170	» 3000 » » » »	5.24
170	» 5600 » » » »	3.83
170	» 48000 » » » »	2.61
170	» 86000 » » » »	2.18
	Sea water	21.5

of the salts from the air is selective it can be inferred that only half of the sulfur in rainwater from this place is derived from the sea by the same mechanism as the chloride; the rest must have been delivered in another way.

The distance from the sea has a great influence on the Cl/S ratio as seen from the dutch values. In France, however, FARCY (105) has found a quite opposite trend, the Cl content being rather constant inland while the S was highest at the coast.

There is a recent investigation carried out in Australia by ANDERSON (13) on the composition of lake waters in arid districts compared to the composition of sea water (see table 9).

Table 9. The relative composition of some lake and river waters in south east Australia: Cl + Br = 100.

	Lake Corangamite		Woody Yallock River	Snarket Creek	Sea water
	Jan. 4 Low water level	June 12 Very low water level			
Cl+Br	100	100	100	100	100
Cl	—	99.84	99.86	99.86	99.85
Br	—	.16	.14	.14	.15
SO_4	2.99	2.89	6.5	8.2	10.26
CO_3	.91	.60	—	1.2	.45
HCO_2	.86	.54	7.1	5.0	
NO_3	—	—	.7	—	—
Ca	.29	.11	9.7	5.5	3.83
Mg	13.09	13.27	36.4	32.1	19.59
Na	86.26	87.00	65.7	74.7	85.17
K	3.95	3.21	3.8	2.4	1.82

The runoff from some of the lakes is very small and practically all their chlorides are derived from the sea, carried inland and precipitated by rain. He found, however, that the relative sulfate contents in these lake waters were less than in the sea; sulfate being lost in some way. Anderson suggested that this loss was due to the reduction of SO_4 to H_2S as there was a definite smell of H_2S around the lakes. It is also seen from the CO_3 and HCO_3 contents that the pH of the waters must have been quite high, apparently due to this reduction process. The loss of sulfate by reduction to H_2S in the lakes must have been very high and proportionally higher than in the sea as it can be expected that the rainwater carried relatively more sulfate than chloride from the sea.

An interesting investigation was carried out by Boss (46) on the south west coast of Africa. Due to a cold water current in the sea reaching the surface at this place there is a permanent fog in this district which is a desert. He found that the fog was relatively richer in sulfur than in chloride compared to sea water and that the proportion of sulfur increased inland. For an explanation he suggested a small sulfur cycle which took place in the following way. Both chlorides and sulfates are carried inland by the fog but the chlorides seems to be leached down into the soil much easier than the sulfates especially as most of the latter are in the form of $MgSO_4$. During dry weather fine particles of $MgSO_4$ are carried from the earth's surface to the air where they serve as condensation nuclei. Very little sulfate is actually lost in this way and the process leads to an enrichment of $MgSO_4$ in the top soil while chlorides are leached down into the subsoil. Perhaps such a small cycle of sulfur is partly responsible for the lower Cl/S ratio in rainwater compared to sea water.

To summarize it can be concluded that a great part of sulfur in precipitation is of oceanic origin either brought into the air by spray or as H_2S formed by reduction of sulfate in the sea. Another source of sulfur in rainwater is coal, sulfide ores and wood which, on combustion, liberate considerable quantities of sulfur. A third source is H_2S liberated from organic matter on decay, and a fourth may be of inorganic litospheric origin.

D. Chloride in precipitation

1. Results from different places

The first determinations of chloride in rainwater were made at the beginning of the nineteenth century but the first more systematic work was not published until 1851 when ARAGO (14) in Paris reported monthly amounts for a half year period. Another systematic investigation was carried out by SMITH (277) who in 1872 published his well known "Air and Rain", where he has given analyses of rainwater collected at different places in the British Isles. Some of the chloride analyses in his work are given in table 11.

One of the longest series of analyses was carried out at Cirencester by KINCH (158, 159) covering a period of 26 years.

The determinations of chloride in rainwater has, of course, been made mostly for other reasons than agricultural. The salt transport from the sea may be of agricultural but is also often of technical value. In meteorology chloride in rainwater is of great interest as it can probably give a good indication of the origin of the air masses.

The amount and composition of chloride in rainwater from different places are tabulated in table 10. The European investigations are not so extensive as the corresponding ones for ammonia and nitrate and the American ones are also few, compared to the sulfur investigations. The New Zealand and Australian investigations are rather extensive because of the great practical importance of the salt transport from the sea by rain in these places.

The amounts and concentrations of Cl in rainwater do not vary very much in inland places whereas coastal places show a great variation. Remarkably high figures are however obtained from Iowa and Tennessee in the U.S.A., places which are really inland situated at great distances both from the Pacific Ocean and the Atlantic. For Iowa it has been assumed that all chloride in rain water is derived from the sea as no other likely sources exist in the neighbourhood.

2. Origin of chloride in precipitation

It was suggested long ago that the sea was the greatest source for chloride in rainwater as rainwater in coastal places contained by far the

Table 10. Chloride in precipitation at different places.

Ref.	Place	Period	Precipitation mm	mg Cl/l	kg Cl/ha
	Europe				
131	Denmark: Askov	1925/26	740	4.2	30.8
131	Blangsted	1925/26	600	4.7	29.3
131	Hornum	1925/26	495	5.8	28.6
131	Spangsbjerg	1925/26	730	12.4	90.9
14	France: Paris	1851	560	2.3	13.0
43	Nantes	1863	—	—	8.4
188	Germany: Müncheberg	1933/34	405	3.6	14.4
158, 159	Great Britain: Cirencester	1874—1900	775	3.2	24.6
213, 254	Rothamsted	1887/88-1915/16	740	2.4	18.0
81	Garforth	1907/08-1908/09	720	3.1	22.0
312	Russia: Leningrad	1909/10	646	3.6	23.0
312	» outside town	1909/10	669	2.4	15.8
312	Pawlowsk	1909/10	522	1.6	8.3
312	Sapolje, Kreise Luga	1909/10	486	3.0	14.6
312	Smolensk	1909/10	660	3.1	20.3
312	Jekaterieslaw	1909/10	339	3.8	12.8
312	Schotilowska, Tule distr.	1909/10	476	1.8	8.7
312	Borowoje, Ssarnara	1909/10	541	2.0	10.6
209	Spain: La Guardia (at the coast)	1892/93	1433	31.2	447
	America				
75	Geneva N.Y.	1919—1928	900	2.0(2)	18
15, 117, 161, 235, 246, 269, 290, 306, 313	Iowa: Mont Vernon (8 years)	1913/14-1931/32	—	—	72
195	Tennessee: Knoxville	1922	—	—	140
195	» Univ. Farm	1922	—	—	70
288	Kansas: Madison	1915—1918	—	—	18
133	Barbados	1886—1889	—	8.1	—
133	British Guiana: Georgetown	1890—1895	—	4.6	—
	Asia				
216	Japan: Tokyo	1938	—	1.8	—
216	Kobe	1938	—	2.3	—
216	Hamamatu	1938	—	2.4	—
18	India: Ceylon	1898/99	—	9.7	202
226	Bombay	1938	—	13.6	—
	Africa				
156	Bloemfontein, South Africa	1910/11-1911/12	550	.9	5.1
156	Cedera, » »	1912	680	2.7	18.1
156	Durban, » »	1911—1912	910	8.1	73.5
156	Grahamstown, » »	1911/12	590	4.5	26.3
	New Zealand				
119	Rangatai 500 m from the sea	1948—1950	965	22.4	216
119	Baring Head 500 » » » »	1948—1950	820	20.2	166
119	Kelburn 6500 » » » »	1948—1950	1220	8.7	106
119	Pirinoa 6500 » » » »	1948—1950	840	11.8	99
119	Belmont Hills 8000 » » » »	1948—1950	1400	7.5	106
119	Levin 10000 » » » »	1948—1950	1090	4.6	50
119	Wallaceville 18000 » » » »	1948—1950	1270	5.8	74
119	Palmerston North 32000 » » » »	1948—1950	1040	4.8	50
119	Martinborough 32000 » » » »	1948—1950	790	7.6	60
119	Te Awa 48000 » » » »	1948—1950	1070	3.3	35
119	Waingawa 61000 » » » »	1948—1950	860	4.8	41
125	Lincoln, Canterbury	1884—1888	750	8.8	66.4

Ref.	Place	Period	Precipitation mm	mg Cl/l	kg Cl/ha
	Australia				
13	South Australia, York Pennisula.....	1923—1937	445	21.6	96
13	Victoria: O'Shamarsy.....	1923—1937	1620	2.7	44
13	East Camberwell.....	1923—1937	715	5.7	40
13	Yarra	1923—1937	1130	4.4	49
13	Upper Murray.....	1923—1937	840	1.6	8.4
13	Barrwan River Catchment..	1923—1937	645	8.4	54
282	West Australia: Merredin.....	1933—1936	260	4.2	11.0
282	Salmon Gums.....	1933—1936	320	5.7	18.4
316	Esperance.....	1926	—	20.7	—
316	Geraldton.....	1926	—	28.7	—
316	Perth.....	1926	—	16.5	—
316	Coolgardie.....	1926	—	12.0	—
316	Cue.....	1926	—	9.6	—
316	Mundiwindi.....	1926	—	3.5	—
316	Rawlinna	1926	—	6.3	—
316	Wilunna.....	1926	—	4.0	—
316	Condon.....	1926	—	8.2	—

highest amounts of chloride. This is also clearly seen in table 11 where SMITH's data are tabulated. Special investigations have also been carried out in order to asses the influence of the sea. Recent investigation from New Zealand (119) shown in table 10 shows very clearly the influence of the sea. Another investigation which is of interest in this connection is that published in 1905 by JACKSON (151). He analysed spring waters in New England and New York and from his analysis he was able to draw isochlors for the whole area. These isochlors show very clearly the influence of the sea in this district. Later CONWAY (78) used his data to set up a mathematical relation between the chloride content in uncontaminated spring waters and the distance to the sea. The relation can be written

$$Cl = 5.7 \cdot e^{-0.0367\,x} + 0.55 \cdot e^{-0.00242\,x} \quad (D)$$

where Cl is expressed in mg/l and x is the distance from the sea in kilometers.

The first term represents a relatively rapid decrease of Cl inland while the second one, although small shows that after the first rapid decrease there is a much smaller decrease.

There is an investigation from Holland carried out by LEEFLANG (177) in 1938 of a similar kind as the New Zealand one although better suited for the calculation of a relationship of a similar type as (D) than the New Zealand investigation. The relationship corresponding to (D) from LEEFLANG's figures reads

$$Cl = 11.7 \cdot e^{-0.525\,x} + 3.0 \cdot e^{-0.0230\,x} + 3.0 \quad (E)$$

Also in this case the relation can be expressed by two exponential factors but it also involves a constant which is reasonable as there is always chloride in rainwater even very far

Table 11. Cl-content in precipitation in Great Britain from Smith's investigations (277).

Place	mg Cl/l
London, spec. for 1869.....	1.2
Birkenhead, Liverpool.....	3.1
Scotland, inland country places.....	3.3
» , near an alkali work.....	3.3
England, inland country places.....	3.9
Manchester, 1869—1870.....	5.7
Scotland, towns (not Glasgow).....	5.7
Newcastle on Tyne.....	7.9
England, towns.....	8.5
Glasgow.....	8.7
St. Helen's.....	9.3
Liverpool.....	9.9
Scotland sea coast country places, west	11.9
Scotland sea coast country places, east and west.....	12.2
Scotland sea coast country places, east	12.6
Runcorn.....	25.0
Waterlow, near Liverpool.....	35.5
Ireland, Valentina.....	47.3
England, sea coast place, west.....	54.6

from the sea. The coefficients in (D) and (E) differ considerably. Firstly the initial Cl content seems to be higher in (E) than in (D) and the decrease in the first term is also much more rapid. The second term in (E) decreases, however, much faster than the same in (D). It is probable that the differences are significant and indicate differing meteorological conditions on the North American east coast and the European west coast.

The experimental points and relation (E) are shown graphically in fig. 8.

An investigation similar to that of Leeflang has also been carried out in France by FARCY (105) at different places on the road from Brest to Paris. As a rule the Cl contents are higher than the Dutch ones with a very slow decrease inland. In one serie the Cl content in rainwater at the sea was 19.0 mg per litre and 18.7 mg per litre at 24.3 km distance from the sea. It is thus seen that the conditions for the transport of chloride from the sea may vary considerably from place to place.

CONWAY (loc. cit.) has discussed the sources of Cl in detail. From equation (D) he calculated by integration over all coast lines that the total amount of Cl brought inland from the sea each year amounted to $0.69 \cdot 10^{14}$ g. Compared to other possible sources he mentions that the total salt production by human activities amounts to $0.09 \cdot 10^{14}$ g per year

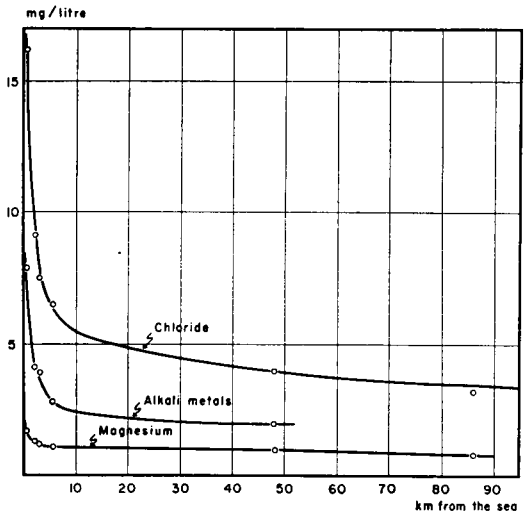

Fig. 8. The concentration of Cl, Mg and Ca in precipitation in Holland as a function of the distance to the sea.

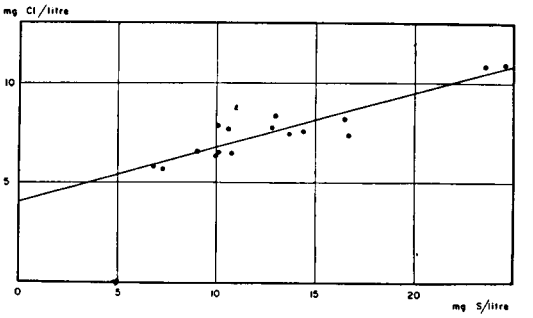

Fig. 9. The relation between the concentration of S and Cl in precipitation around Leeds, Great Britain.

while human secretion amounts to $0.05 \cdot 10^{14}$ g. Combustion of coal contributes only $0.015 \cdot 10^{14}$ g per year. From equation (D) and the French investigation mentioned it is obvious that the figure $0.69 \cdot 10^{14}$ g Cl per year may be far too small; obviously other sources of Cl in rainwater become insignificant compared to the sea.

In industrial places the Cl content of the rain may rise somewhat but compared to the increase in sulfur the increase in Cl is smaller. This is seen from an investigation by CROWTHER & STUART (82) who collected rainwater at different points around Leeds in England. From their data on S and Cl it is possible to get a linear relation between these two which reads

$$Cl = 4.06 + 0.275 \qquad (F)$$

This relation is shown diagrammatically in fig. 9. The correlation coefficient is equal to 0.925. It is seen that the increase in Cl is about one fourth of the increase in S due to atmospheric pollution. In other places it may be even less.[1]

Equation (F) is interesting as it shows that in absence of any industrial contamination the Cl content of rain should amount to 4.1 mg pro litre, a very reasonable figure for England (Cf. tab. 11 inland country places in England with 3.9 mg Cl pro litre).

In their final report of the Rothamsted investigations RUSSEL & RICHARD (254) mention that the Cl content in rainwater at

[1] The increase of Cl in this case can hardly be due to combustion of coal as the chloride content of coal is very low. It is probably connected to some chloride containing chemicals used in the industry.

Table 12. Cl in precipitation near volcanoes.

Place and date of sampling	Ref.	pH	Cl mg/l
Vesuvius, Italy			
March, 5, 1934	47	2.96	247
» , 17, »	47	3.16	193
May, 10, »	47	2.39	202
» , 18, »	47	2.82	105
June, 30, »	47	3.09	88
Paracutin, Michoacan, Mexico 1947 12 km N of the foot of the Volcano	55		440

Rothamsted had a tendency of increasing with time, a fact they attributed to the increased combustion of coal. In the Cirencester series (KINCH, 159) it is, however, impossible to detect any such trend.

Another source not mentioned before is Cl in volcanic gases. There are a couple of investigations which may be of interest in this connection. One is carried out near the Vesuvius volcano by BOTTINI (47) in 1934. The composition of 5 samples with respect to pH and Cl is shown in table 12, together with an analysis from the Paracutin volcano in Mexico (BULLARD, 59). It is seen that the chloride figures are very high and, further, that in the Vesuvius' figures at least part of the Cl is present as HCl as the pH is very low.

It is evident that some chloride may locally be of volcanic origin but the total contribution by volcanoes is insignificant compared to the contribution by the sea.

3. Meteorological factors influencing the Cl-content of rainwater

Some of these factors are obvious from the preceding discussion but as they have been pointed out in the literature they will be discussed here.

a. *Rainfall*

As in the case of other compounds there is generally an inverse relationship between the Cl-content and the amount of rainfall. As examples it can be mentioned that MILLER (213) using monthly averages calculated for Rothamsted the relationship to be approximately

$$Cl = 2+3/p$$

in winter

$$Cl = 0.5+3/p$$

in summer and

$$Cl = 1.25+3/p$$

for the whole year. In these expressions p is the monthly rainfall in inches.

The expressions indicate that there is a lower limit of concentration in the precipitation, and this limit is higher in winter than in summer.

SUGAWARA et al. (281) have recently (1946—47) analysed rainwater in Japan and found at Nagoya (4 km from the sea) also an inverse relationship between rainfall and Cl-concentration.

Also in a few other investigations an inverse relationship can be traced.

b. *Wind direction and wind strength*

It is a well known fact that near the sea the Cl concentration in rainwater is largely influenced by the wind direction at the time of rainfall. Also in inland places the Cl-concentration may be influenced by the wind direction, a fact which at least to a certain extent can give information as to the origin of the air masses.

In Leningrad WITUYNJ (312) found that the Cl content was higher in spring than in autumn depending on the westerly winds prevailing in the spring. In Denmark HANSEN (131) found that the Cl content even at inland places was highest during westerly winds. In Donnersberge in Germany MENZL (207) found that the Cl content of rime frost deposited under different wind directions was the following: N, 8.01; NE, 4.78; E, 3.43; W, 9.67 and NW, 10.53 mg/l. This shows that the air masses from north to west carried most Cl. In snow he (208) found about the same while rain from the west had the highest Cl concentration. In Haid also in Germany rain from SW had the highest Cl concentration followed by rain from NW. In Hohen Sonnblick in Austria at 3106 metres' altitude LAUSCHER & SCHWARZ (170) found that winds from north carried less chloride than winds from the south.

From this is seen that at lower altitudes the sea has still a noticable influence on the Cl

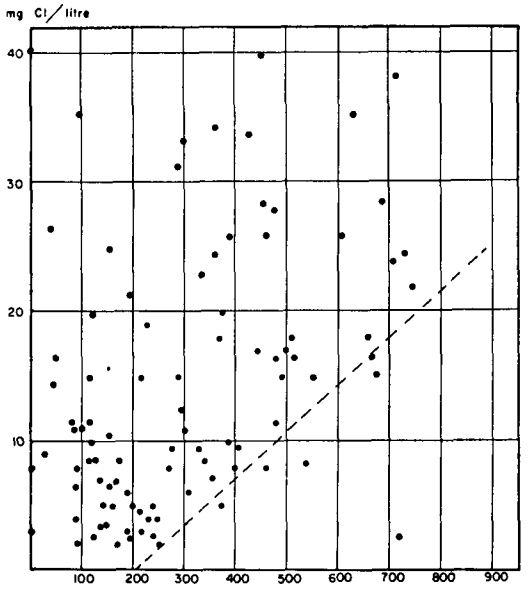

Fig. 10. The relation between the concentration of Cl and wind length at Perth, West. Australia.

concentration of atmospheric precipitation even in places far from the sea. At higher altitudes this influence is not so pronounced.

The wind strength when the winds come from the sea is reported to have some influence on the Cl content of rain. MIYAKE (216) in Japan found a strong correlation between Cl content and wind strength. WOOD & WILSMORE (316) in their research on the salinity of rain in Western Australia correlated the Cl content of rain at Perth with the total way the wind had travelled from the sea in 24 hours. It is a rather interesting relationship although the correlation is not very strong. In fig. 10 the Cl content of the rainfall has been plotted against their computed wind length sea wind in miles/24 hours.

From fig. 10 it is seen that the Cl content of the rain is independant of the wind strength up to a wind velocity of 200 miles pro 24 hours with a lower limit of 2 mg Cl pro litre. Above that velocity it can be stated that in general

$$Cl \geqq -7.2 + 0.036\, v \qquad (G)$$

where v is the wind length in 24 hours, this lower limit being indicated on the figure by the dashed sloping line. Deviation from this rule can, of course, occur on single occasions. The interpretation of this rule is simply that at high winds one cannot expect low Cl contents in the rains.

c. *Altitude*

There is an old investigation from France carried out by BOBIERRE (43) in Nantes in 1863 which may show differences at lower altitudes. He collected rainwater from two heights, 7 m and 50 m above the ground and analysed them. In the yearly averages there was no difference in the Cl contents but in winter the samples collected at 50 m contained more Cl than those collected at 7 m while the reverse was true during summer. The reason for this may be local.

There are, however, several investigations made at different altitudes where precipitation has been collected on mountains.

Analyses of rime frost on the Hohen Sonnblick in Austria made both by LIPP (192) and LAUSCHER & SCHWARZ (170) gave relatively low Cl values. LIPP found on 2962 m altitude an average of 1.1 mg Cl/l for 12 determinations with variations from 0.25 to 5.90. LAUSCHER & SCHWARZ found on 3106 m an average of 0.21 mg for 15 determinations with variations from 0.035 to 0.554 mg pro litre. In *Donnersberge* (207) on 853 m altitude the average for 17 determinations was, however, as high as 7.23, rain and snow containing about the same. Some analyses of water from lakes and rivers in the Haldde district in northern Norway presented by KÖHLER (167) indicate that the Cl content in general decreased with increasing altitude although there were variations, according to Köhler probably due to topographic conditions influencing the turbulence of the air.

The Cl content of precipitations thus seems to decrease with increasing altitudes. This has also been predicted by KÖHLER (166) from theoretical considerations.

d. *Seasonal variation*

The seasonal variation in the Cl concentration of precipitation seems to be entirely connected to the prevailing winds as can be expected and also to the general meteorological conditions such as wind strength. Such observations are reported from several places.

4. The "Köhler" distribution of Cl-concentrations

In 1925 KÖHLER (165) published a report on an investigation of the Cl concentration in white frost formed at high altitudes by precipitation and freezing of undercooled water particles in clouds. This type of frost will be called cloud frost here. From 49 analysis of collected cloud frost he found that the Cl concentration in the melted frost could be expressed by

$$Cl = 3.523 \cdot 2^n \text{ mg pro litre}$$

where n is a positive or negative integer. Of course there was some variation within each group but the group formation was quite obvious and statistically significant.

The distribution of Cl concentrations will be called the "Köhler" distribution here. It is of interest as he had earlier found that the masses of cloud particles also showed a group formation which could be written

$$m = B \cdot n$$

where B is a base number and n so far has been shown to take the values 2, 3, 4, 5, 6, 7, 8, 10 and 12.

A surprising fact from this frost analysis was, however, that no correlation between the masses of the cloud particles and their Cl concentrations was found.

The explanation offered by Köhler for the observed Cl distribution is that the weight of the spray particles, which leave the sea is distributed in the same way as the Cl concentrations, but the reason for such a distribution in the spray is, of course, difficult to give.

On basis of further investigations in the same place (the Haldde district in northern Norway) he found that the most common particle size had a radius of $8.82 \cdot 10^{-4}$ cm. As the most common Cl concentration was 3.523 mg/l the most common amount of sea salts in a particle is $1.847 \cdot 10^{-14}$ g but that smaller or greater amounts could occur. For comparison it can be mentioned that ALIVERTI (6, 7) in an investigation from Pavia in Italy found that the average amount of salt in condensation nuclei was $0.075 \cdot 10^{-14}$ which corresponds to a value of n in Köhler's formula between —4 and —5, if the particles had the same masses.

Investigations similar to Köhler's were later carried out by LIPP (192) and LAUSHER & SCHWARZ (170) in the Hohen Sonnblick and by MENZL (207) in Donnersberge. The data of LIPP and LAUSCHER & SCHWARZ are, however, too few to detect any "Köhler" distribution. The Cl values from Donnersberge are surprisingly high, varying from 3.43 to 15.33 mg Cl/l.

There are some other investigations of Cl in rainwater where distinct "Köhler" distributions of Cl concentrations are found. In these the rainfalls are, of course, analysed separately. In *Leiden* in Holland (4 km from the sea) ISRAËL (150) found that the Cl content of rainwater could be expressed by

$$Cl = 3.42 \cdot 2^n \text{ mg pro litre}$$

where $n = 0$ and 1 seemed to be the most common values.

The West Australian investigation mentioned before (WOOD & WILSMORE 316) show also weak signs of the "Köhler" distribution as well as MENZL's (208) on rainwater from Donnersberge.

In an investigation from Japan MIYAKE (216) also found "Köhler" distributions of Cl concentrations in rainwater although the base numbers varied somewhat. In Tokyo he found

$$Cl = 2.10 \cdot 2^n \text{ mg Cl/l}$$

and in Kobe

$$Cl = 1.89 \cdot 2^n \text{ mg Cl/l}$$

and in Hamamatu

$$Cl = 4.0 \cdot 2^n \text{ mg Cl/l.}$$

The most pronounced "Köhler" distribution of Cl contents in rainwater is found in the analysis from Mont Vernon in Iowa, U.S.A. The investigations are extended over 10 years with analysis of separate rainfalls. The analyses are, however, apparently carried out by different workers under the supervision of N. KNIGHT at the Mont Vernon University (For references see 15, 117, 161, 234, 246, 269, 290, 306 and 318). In most of the reports only the total amount of Cl/acre is given. The author has therefore recalculated the Cl concentrations from these amounts and the listed rainfalls.

Table 13. The distribution of Cl-concentrations of the rainfalls at Mont Vernon, Iowa. Cl = 0.355 · *s*.

s	1	3	7	8	10	13	14	15	16	18	20	22	23	24	25	26	27	
Frequ.	1	1	1	3	30	1	2	3	7	2	94	4	1	5	4	5	3	167
s	28	29	30	32	33	34	35	36	40	43	45	48	49	50	52	60	70	
Frequ.	1	1	71	2	1	2	3	5	14	1	3	1	1	1	1	4	2	114
s	73	88	92	140														
Frequ.	1	1	1	1														4
Total Frequ.																		285

From the total number of analysis it can be concluded that the analyses of Cl were carried out with a precision of 0.355 mg Cl pro litre as practically all the figures can be represented by the formula

$$Cl = 0.355 \cdot s$$

where *s* is a positive integer ranging from 1 to 140. The number of Cl analysis for different *s* is shown in tab. 13.

In connection with KÖHLER's work it is of interest to note the high frequency of *s* equal to 10, 20 and 40. These are well represented by Köhler's formula with the base number 3.5 and *n* equal to 0, 1 and 2. On the other hand the high frequency of the figure 10.65 (*s* = 30) indicates a deviation from the "Köhler" distribution, 10.65 being the sum of $3.55 \cdot 2^0$ and $3.55 \cdot 2^1$.

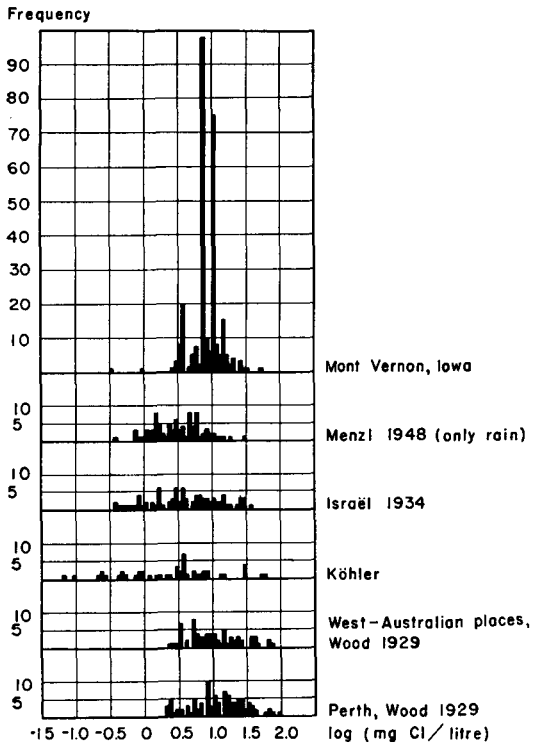

Fig. 11. The distribution of log (mg Cl/l) for some places.

The distribution of log (mg Cl pro litre) for Perth and some other places in Australia (WOOD & WILSMORE, 316) Haldde (KÖHLER, 165) Leiden (ISRAËL, 150) Donnersberge and Haid (MENZL, 208), and from Mont Vernon, Iowa is shown in fig. 11. The intervals have been taken equal to 0.05 units. It is seen that the Haldde series shows the widest distribution and very distinct frequency groups. This is also quite distinct in the Leiden series as well as in the Donnesberge-Haid series. In the Mont Vernon series this frequency group formation is very pronounced. The Australian series have, at least, a tendency of frequency group formation. The base values for this series seems, however, to differ from the European and American series, a fact which is of interest in connection with the earlier mentioned Japaneese observations.

A frequency group formation can also be traced in the figures published by HEYMANN (140) from Oosterveld in Holland which has been mentioned earlier. On the other hand the Zeeveld figures do not show any such tendency. As Zeeveld is close to the sea the figures may be influenced by sea spray which has precipitated straight into the rain gauge.

It should be mentioned that HOUGHTON & BENRIS (142) did not find any frequency group formation in the Cl values from cloud water collected on Mount Washington. However, they did not give any details of their work.

Table 14. See the text.

Catchment area	Area km²	Rainfall mm	Ratio Discharge to Rainfall	Mean conc. of Cl. Rain-water	River water calc.	River water Actual
Upper Murray, Vic.	26600	842	0.283	1.0	3.5	3.4
O'Shannany River, Vic.	121	1620	0.520	2.7	5.2	4.75
Yarra River, Vic.	2530	1110	0.311	4.4	14.2	15.3
Barwon River, Vic.	3710	642	0.075	8.4	112	134
Helena River, West. Austr.	1480	713	0.034	6.7	197	203

From all the results shown it seems quite evident that a frequency group formation of the Cl content in precipitation does exist.

5. Practical importance of the transport of oceanic salts inland

In arid places the transport of sea salts inland has a very great practical importance. In western Australia the transport of sea salts inland has caused an increase in the salt content of the soil, the streams and the water reservoirs (WOOD, 315). In 1914 WELLER (303) found that the Mundering reservoir catchment area (about 1400 square km) received around 90 kg of oceanic salts pro hectare and year and as there is practically no outlet from this area to the sea the salinity of the reservoir water has been steadily increasing. WOOD (315) also mentions that in many of the water reservoirs for the railways in Western Australia the salinity of the water had increased to such a degree that the water could not be used for the locomotives. This process has been enhanced in many places by the clearing of land from natural vegetation thereby causing a rapid runoff of water to the reservoir, the runoff water carrying the accumulated salts in the soil.

A more recent Australian investigation mentioned earlier (ANDERSON, 13) concerns arid parts of the State of Victoria. He found from analysis of river waters from different catchment areas that, knowing the rainfall, runoff and the salt content of the rainfall, it was possible to calculate the composition of the river waters. Comparison between calculated and found values shows a very good agreement. (See table 14.) As mentioned earlier he also found that the composition of some lake waters in this district was very similar to that of sea water.

It is obvious that under arid conditions where the percentage evaporation is high an accumulation of salts must occur in the soils. This is, no doubt, a very common cause of saline soils in desert and semidesert district although some saline soils may be due to salt lenses in the soil at greater depths and a high evaporation.

The salt transport seems to be of some importance also in New Zealand (119) where, in some places, the soil processes appear to be effected by the accumulation of oceanic salts.

A concentration of oceanic salts in the soil water seems also to take place in humid areas. WARINGTON (295) published an account of analysis of drain and well waters in Rothamsted in 1887. He found that the Cl content of drain water from unfertilized plots was higher than in rainwater. This, he explained, was due to the evaporation which takes place during summer. This fact suggests an indirect method of estimating loss of water by evaporation from uncultivated soils. This, however, is possible only where no well waters containing Cl can reach the upper layers of the soil.

E. Iodine in precipitation and air

The presence of iodine in atmospheric precipitation is mainly of medical interest in connection with goitre and most of the more recent investigations seem to have had, as a purpose, the making of a survey of existing sources which can deliver iodine to the air.

As early as in 1852 MARCHAND (197) showed that rain, snow and hail contained considerable amounts of iodine and bromine. At about the same time CHATIN (71) found in analysis of rain and spring waters from Guiana in South America 5 μg iodine/l and in Nice 4 μg/l. In an investigation from Pisa around 1860 DE LUCA (193) found iodine both at 54 and 18 m

height above the ground. (The samples were taken from the famous leaning tower.) On the other hand CHAIRY (70) in 1884 did not find any iodine in rainwater collected in Algiers despite the fact that the content of oceanic salts in the water was quite high.

More systematic work was started by VON FELLENBERG (107, 108) who in 1926 published a paper about the iodine cycle in nature. Most of his work is concerned with analysis for iodine in the air.

In 1927 HEYMANN (140) in Holland published analyses of air, rain and drainwater. A few air analyses from Leiduna (6 km from the sea) in 1924—25 are given below.

Wind direction	E	SW	SW	E	SO	SSW	NW
μg I/l	0.5	1.0	0.2	1.5	0.1	0.05	0.2

The rain analysis were carried out in Oosterveld and Zeeveld. He found an average of 3.6 μg for Oosterveld and 3.7 μg for Zeeveld. The distribution of the values are given below

	μg I pro litre	1	2	3	4	5	6
Frequency at	Oosterveld ...	5	0	7	7	5	1
	Zeeveld	3	6	3	4	8	1

	μg I pro litre	7	8	9	10	11	12
Frequency at	Oosterveld ...	3	1	1	0	0	0
	Zeeveld	1	2	1	0	0	1

There is as seen no difference in the iodine content of rain from these two places. The average Cl content in Oosterveld was 12.6 and in Zeeveld 28.8 mg Cl/l, Zeeveld being at only 0.4 km distance from the sea. Further there was no correlation between the Cl and the iodine figures. This suggests a different mechanism by which iodine is brought from the sea which in this case was the only possible source.

Further the ratio Cl/I in the samples was very much lower than in sea water as sea water contains only 20 μg/l. HEYMANN further observed a great difference in the behaviour of Cl and iodine. From his drain water analysis he found that while Cl was higher in the drain water than in the rain (due to evaporation) the reverse was true for iodine. A loss of iodine must have occured in the soil. VON FELLENBERG (108) found a little later that water containing iodide lost iodide when in contact with air. He suggested that iodide is oxidized to iodine which is volatile and escapes from the water to the air. Further he found that the air in spas which used iodine rich ground water contained a high concentration of iodine, up to 5 γ pro cubic metre, a fact which supported his theory.

In a series of investigations CAUER (63, 64, 65, 66) continued VON FELLENBERG'S work. He also analysed air at high altitudes 3000 m for iodine and found about the same concentration as near the ground, i.e., it varied between 0.1 and 1.4 μg/cubic metre. This is also different from Cl which seems to decrease at higher altitudes.

CAUER developed a theory that the oxidation of iodide to iodine was done by ozone, present in the air, and he showed that ultraviolet radiation of iodide containing water caused great losses of iodine. The iodine cycle in nature is thus more complicated than the chloride cycle.

The sea is thus a great source of iodine which is partly brought to the air by spray and partly by oxidation of iodide to iodine. Iodide carried to the soil by rain may also be brought to the atmosphere by this oxidation.

An important source of iodine in air in the western Europe is iodine released in the process of burning seaweed as practiced to a great extent in Bretagne in France. Seaweed contain rather great amounts of iodine and Cauer estimated that about 6 per cent was lost during the burning process. In 1933 and 1934 this amount was equal to 65 tons a year, an amount which he considered to be sufficient to raise the iodine content of the air in western Germany up to 2 μg pro cubic metre.

Another source is from coal used in industry but this contribution is very small.

F. Other inorganic compounds in precipitation

Occasionally rain and snow has been analyses for other compounds than those discussed. Some old analyses are given by PIERRE (237), ARAGO (14), BARRAL (20), DE LUCA (193) and ROBINET (248). Of these that of ARAGO is of greatest interest. He collected rainwater in Paris from July to December 1851 and analysed for Ca and Mg. The average for this time is 6.5 mg CaO and 2.1 mg MgO/l.

More recently MAC INTYRE & YOUNG (195) made analysis for Ca, Mg, K and Na in rainwater collected at some points in Tennessee in

U.S.A. The atmospheric pollution was, however, quite strong at these places due to industrial activities.

In Europe some recent data on Ca and Mg in rainwater have been given by FARCY (105). He collected rainwater in the neighbourhood of Brest in France. He concluded from his analysis that part of Ca and Mg in the rainwater was of litospheric origin. FARCY also found Mn in the rainwater on some occasions. He believed that it had been carried over the Atlantic from America.

In a series of papers from 1943 to 1947 BERTRAND (29—35) gives some data on Ca and Mg in rainwater collected at Grignon and at Paris in France. In Paris he found as an average a Mg/Ca ratio of 0.089 (the soluble part) and in Grignon 0.049. The same ratio in sea water is 3.16. Most of the Ca must then have been of litospheric origin while at least part of the Mg was derived from the sea.

In Holland LEEFLANG (177) analysed rainwater collected at different distances from the sea also for Ca and Mg. The results are shown below

Distance from the sea in km	0.44	2.28	3.0	5.6	48.0	86.0	Sea water
Ca mg/l	2.5	1.9	1.7	1.7	1.7	1.7	
Mg ,,	1.7	1.3	1.2	1,1	1.0	1.8	
Mg/Ca	.68	.68	:70	.65	.59	.47	3.16
Mg/Cl	.105	.143	.160	.169	.250	.250	.068
Ca/Cl	.154	.209	.226	.262	.425	.532	.021

and are shown graphically in fig. 8.

It is seen that Mg decreases regularly inland whereas Ca remains comparatively constant. The Mg/Cl ratio increase 2.5 times inland but is of the same order of magnitude as in sea water near the coast. The ratio Ca/Cl is much higher in the rainwater than in the sea even near the coast. It is about 25 times higher far from the coast.

A recent work from Japan gives similar results as those of LEEFLANG. From rainwater analyses during 1946—49 SUGAWARA et al. (281) reported the following ratios.

Place	Sugashima	Nagoya	Matsumoto	Sea water
Distance from the sea in km	0	4	100	
Na/Cl	.05	.55	.58	.55
Mg/Cl	.080	.109	.344	.068
Ca/Cl	.138	.172	3.76	.021

The Na/Cl ratio is remarkably constant but the Mg/Cl and Ca/Cl ratios shows about the same trend as mentioned before.

SUGAWARA et al. offer an interesting explanation for the variation in these ratios. They regard the salts in rainwater as derived from the sea but they are washed down selectively, the most hygroscopic salts being washed down near the coast. Salts like $CaSO_4$ which are non-hygroscopic can be carried far inland, thus raising the Ca/Cl ratio considerably as chlorides of Mg and Ca are much more hygroscopic than corresponding sulfates and are precipitated near the coast. They found a similar trend in the ratios in a vertical direction by analysing precipitation, rain and snow from different altitudes. Also LIPP (192) found a considerable enrichment of Mg over Cl in rime frost on Hohen Sonnblick in Austria.

BERTRAND (36, 38, 39) also determined K in some rainwater samples collected in Paris. He found 0.22 mg K/l in 1942 in a sample and 0.42 in another. The corresponding Mg figures were 0.19 in the first sample. The potassium is thus relatively high. Part of it may be derived from coal by combustion.

In a recent work INGHAM (148) found that vegetation can adsorb comparatively large quantities of Ca from air which is washed down by rain. It is possible that non-hygroscopic salt can be adsorbed from the air in this way and thus be brought to the soil.

A few other compounds have been reported occasionally in rainwater. Small amounts of P_2O_5 have been found (MOORE et al., 217; TRIESCHMANN, 290; DE LUCA, 199). INGHAM (148) found in rain 0.34 mg P_2O_5/l and in drip from tree leaves during rain 2.0 mg/l. These figures are indeed very high compared to earlier estimates.

There is also an analysis on As in rainwater reported by XHORIS (317). The rainwater was collected near Liege and contained 3.07 mg As pro 100 g dry matter from the rainwater. He suggested industrial origin.

G. Miscellaneous

Atmospheric precipitation invariably contains insoluble substances of different origin. These have not been studied very much; on a few occasions with exceptional high contamination they have been investigated mostly in order to determine their origin.

Volcanoes often give rise to local contaminations. Sometimes the dust from eruptions can travel long distances. A well known example is the dust rain which occurred in northern Europe in 1875 when the volcano Askja on Iceland was in action. The distribution of ash and dust from this occasion has been reviewed by THORARINSSON (285).

In the Mediterranean coloured rain may occur. This is due to solid particles from arid regions. By the action of wind the particles are brought up into the air and can be carried long distances before they are precipitated by rain. MENIER (210) found on one occasion in Palermo in Italy that a sample of such dust contained 59.14 per cent sand, 23.91 per cent $CaCO_3$ and 8.58 per cent clay. He suggested as origin the Sahara Desert.

A real dustrain occurred in 1892 in May (NORDENSKIÖLD, 229, 230), the rain falling over parts of Germany, Denmark, Sweden and Finland. It was brought down in appreciable quantities in a comparatively narrow strip from northwestern Germany to Finland. The dust consisted mainly of mineral fragments.

The radioactivity of precipitation has also been investigated. It is reported (77) that snow has an induced radioactivity which disappears after a couple of hours.

Erratum

In part I of this paper, page 231, the eleventh row below the subheading no. 8 stands "cm^3". Should read "cm^2".

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