Air Borne Salts and the Chemical Composition of River Waters

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Abstract

The yearly discharge of chloride by rivers in Scandinavia and in the eastern USA is shown on two charts. The isochlor pattern strongly indicates the marine origin of chloride. From the composition of river, lake and ground water compared to rain water it is inferred that the salt in these are to a very great extent air borne.

Some years ago JOEL V. ERIKSSON (1929) published a report on the chemical denudation in Sweden as estimated from the chemical composition of river waters and yearly runoff figures. This investigation was undertaken by the Hydrological Bureau of the Swedish Institute of Meteorology and Hydrology. They collected water samples for chemical analysis at frequent intervals during a number of years at selected points in our Swedish river systems. In the paper by Eriksson the mean chemical composition and the mean yearly runoff are given, as well as the drainage area for each point. It is thus possible to calculate, for each of the constituents investigated, the mean yearly amount carried into the sea from the different areas. Eriksson divided these amounts into a few classes, represented on the charts by different degrees of shading of the areas. Another way of representation is to let the yearly amounts be represented as point values in the drainage area. This has been done here for chloride and the result is shown in fig. 1. The calculations were done in the following way. Firstly, the percentage Cl of the total inorganic content was calculated and from the data on the daily discharge of inorganic compounds, the daily discharge of Cl. (When given, the planemetered data of the total inorganic discharge were used.) The daily discharge of Cl was then multiplied by 365 and divided by the runoff area.

The positions of the points are not always coincident with the centroid of area due to the use of weighting factors dependent on the distribution of precipitation (cf. WALLÉN 1951). On the Swedish west coast an additional weighting factor depending on the distance from the sampling point to the sea has been employed. Only in one case, however, has this latter factor influenced the position, namely, in the case of the Viskan river drainage area where the sampling station was relatively close to the river mouth.

For some areas no measurements of yearly runoff existed at the time the report was published; values for these regions were estimated by Eriksson. Whenever possible these estimates have been checked against more recent data (MELIN 1954) and corrected if necessary. The rivers for which corrections have been made are:

1. *52-1142 and 1143, Himmelsberget Östra and Västra*

Original calculations were based on an average runoff of 13 l $\sec^{-1}$ km^{-2}. An adjacent river has a discharge of about 10 l $\sec^{-1}$ km^{-2}; this value has therefore been adopted in the present calculations. In addition, there is a misprint in Eriksson's paper (l.c.) with reference to Himmelsberget Västra; the mean amount of inorganic compounds in the runoff is given as 0.041 tons day^{-1}, but a calculation using the given figures for the mean concentration and the runoff estimate yields 0.026 tons day^{-1}.

2. *67-421 Frinnaryd*

The mean runoff is estimated as 9 l $\sec^{-1}$ km^{-2}. It is necessary to reduce this figure by 50 per cent since the whole river has a mean runoff of only 6 l $\sec^{-1}$ km^{-2}.

Two or more sampling points exist on some of the rivers. This circumstance makes it possible to subdivide the total drainage area into two or more regions; for example, the chloride amount measured at an upstream station may be subtracted from that determined farther downstream to yield a figure which is representative of the region lying downstream from the first station. This has been done for the Lagan river (98-208 Lagan and 98-221 Knäred 2), river Klarälven (108-274 Edebäck and 108-279 Skåre) and the river Gullspångsälven (108-240 Timsbron and 108-242 Åtorp). In the case of the Nissan river the upstream area (101-1054 Lövrödjan) is so small that no recalculation for the downstream area is necessary. For the river Närkes Svartå only the observation from the station 61-136 Karlslund has been used, as this sampling point represents practically the largest drainage area of this river.

The following rivers are not shown on the map for various reasons:

1. *67-818 Risbro*

River Dummeån, draining into Lake Vättern. The yearly amount of chloride is fairly high, amounting to 38.4 kg ha^{-1} $year^{-1}$.

2. *108-1024 Alstern*

A small lake region draining into the Timsälven-Gullspångsälven river system on the northeast side of Lake Vänern, with chloride drainage of 17.2 kg ha^{-1} $year^{-1}$.

3. *108-1026 Laxbäcken*

A small river draining into the same river system as in (2), with chloride runoff of 24.2 kg ha^{-1} $year^{-1}$.

4. *108-979 Svenningstorp*

The headwaters of river Lidan close to the southwest shore of Lake Vättern. Chloride runoff of 37.2 kg ha^{-1} $year^{-1}$.

These values of chloride runoff agree fairly well with the figures shown on the map of fig. 1, with the exception of 67-818 Risbro.

Bifurcations of rivers occur at two places in northern Sweden. The estimated drainages for each branch as given by Eriksson (l.c.) are used for calculation of the chloride drainage.

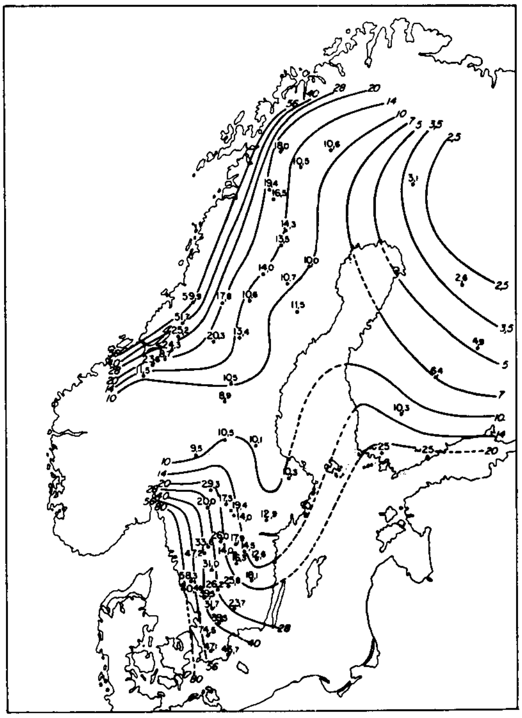

Fig. 1. Discharge of chloride by Scandinavian rivers in kg ha^{-1} $year^{-1}$. Isochlors logarithmically spaced. Note: The figure 74,5 in the southermost part of Sweden should read 52,0.

Data from Norway and Finland are also shown in fig. 1. The Norwegian data, published by Braadlic (1931) cover a rather important area and provide valuable information about the west coast of Norway. The Finnish data are obtained from a recent paper by Viro (1953) on the loss of soil nutrients in Finland.

Looking at the plotted values it is immediately seen that there is a general strong decrease in the values (given in kg ha^{-1}) from west to east. For the analysis of this map logarithmically spaced isolines have been chosen, and it is seen that the spacing between the isolines is remarkably constant in some areas. This would be expected if the chloride was airborne in westerly to southwesterly winds and the rate of precipitation of chloride was proportional to the concentration in the air. As a matter of fact the picture in fig. 1 is remarkably similar to that of chloride in rainwater (cf. Emanuelsson et al. 1954 p. 262) which is shown in fig. 2, redrawn from the original data, at least for southern Sweden. The obvious conclusion is that the chloride in river water has mainly been precipitated from the atmosphere. Still, the amounts in rainwater are appreciably smaller than those in river water, a discrepancy which needs an explanation though some of it may be due to the yearly fluctuations in the Cl precipitation which, as Rossby & Egner (1955) has shown may be considerable.

The contribution of Cl to the river water from the weathering of our bedrocks must certainly be very small. A recent and very interesting survey on the chloride content of bedrocks has been done by Behne (1953) in which a number of igneous and sedimentary rocks have been investigated as well as separated minerals. The estimated average content in both types of rocks were very similar, namely 150 and 154 g ton^{-1}. There are rather great variations mostly depending upon the mineral composition. Most of the chloride is chemically bound, especially in apatites, biotites and hornblendes, and is released by the weathering of the minerals. A minor part is occluded in the minerals and this part is easily removed by fine mechanical division. Now, these amounts are very small anyway and cannot possibly contribute anything noticeable. If, for instance, 15 kg ha^{-1} should be taken from the rock about 0.4 cm has to be completely weathered. On an average such a weathering would release about 1,000 kg ha^{-1} of calcium which also would be found in our river waters whereas Eriksson (l.c., 1929) found an average of 10—20 kg in the granite areas. Thus, possibly one or two percent of the Cl in fig. 1 can have come from bedrock.

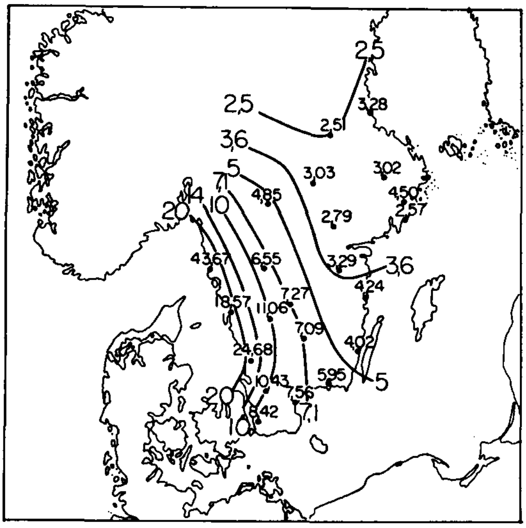

Fig. 2. Chloride in precipitation in southern Sweden in kg ha^{-1} year^{-1}. Isochlors logarithmically spaced.

As parts of Sweden at the end of the last glaciation were submerged, inclusion of chloride into the sedimentary deposits took place and one would expect a certain contribution from this chloride to the river water. On the map in fig. 1 there is, however, no indication of any substantial contribution from earlier submerged areas.

Considering other possible sources, the use of potassium fertilizers in agriculture during the period studied by Eriksson can have furnished on an average over the whole of Sweden 0.5 kg ha^{-1} year^{-1}. In agricultural districts like the southernmost province this gives a contribution of about 5 kg ha^{-1} year^{-1} which is still small compared to the actual values. Further north the drainage basins include large areas of uncultivated land and reduces the contribution of fertilizer chloride in the runoff water correspondingly.

An appreciable part of the Cl in river water must be precipitated from the atmosphere in some other way than by rain and snow. Part of the discrepancy can be attributed to the fact that precipitation like rime, dew and fog water are not represented in precipitation analyses and it is known that the concentrations of salts in such forms is very high (cf. Eriksson 1952). Further the efficiency of the collecting funnels is certainly low for very fine drizzle, which presumably is rich in Cl. There is, however, also the possibility of direct catch of oceanic salts in small particles by obstacles offered by the vegetation to moving air.

The oceanic salts, from which the chloride is derived, occur in the air as very small hygroscopic particles, so-called condensation nuclei. The settling rate of these is insignificant due to the turbulent character of the air but they can be caught from moving air by small obstacles. Such suitable obstacles are grass leaves, spruce needles and similar small objects. The efficiency of catch is a function of wind velocity and the radius of the obstacle, increasing with increasing wind and decreasing radius. As the small particles are hygroscopic they are generally wet, and once they have been caught they can hardly be removed by the wind. Further, as the air near the ground is depleted, more particles will be transported by turbulent exchange from higher layers; this transport can be very effective especially under strong wind conditions over rough surfaces. A spruce or pine forest is probably the most effective device for this accretion of atmospheric salts which, of course, will be washed down by rain. Tamm (1953 p. 88—90) has recently shown that in eastern central Sweden rainwater collected underneath trees contain much more Na and Ca than when collected in the open (Cl was not determined). In one case after a dry period (l. c. table XX) the concentration of Na in the rainwater collected under the outer parts of a spruce tree crown was five times greater than in the rainwater collected in the open. When the rainwater was collected in the middle of a rain period there was no difference.

The order of magnitude of this nucleus capture by vegetation can be estimated in the following way. Only the "giant" condensation nuclei with radii greater than $1\ \mu$ (micron) are large enough to be caught efficiently by spruce and pine needles under the prevailing wind conditions (cf. Woodcock & Gifford 1949, fig. 12). Recent unpublished investigations show that the chloride content of these nuclei in the north of Sweden is of the order of magnitude of one microgram of chlorine per cubic meter of air. If these nuclei are caught by vegetation a density difference of that order would be set up between the "surface" of the vegetation and the moving air. The amount of evaporation of water from vegetation is of the order of some hundred mm per year, i.e. 10^6 kg ha^{-1} $year^{-1}$. The water vapor density difference between the vegetation "surface" and the air must be of the order of 1 g m^{-3}. Therefore, since the transport mechanism for water vapor is essentially the same (though in the reverse direction), the transport of chloride to the vegetation in the condensation nuclei considered here will be of the order of magnitude of several kg ha^{-1} $year^{-1}$. This order of magnitude agrees well with the amount of chloride required to give a balance between deposition by rain and observed runoff, so the mechanism appears to be a reasonable one.

The plant ecological importance of such a direct transfer to the ground and vegetation of atmospheric salts need not be stressed.

As to details in fig. 1 it is seen that the south of Sweden, even on high ground, receives a considerable amount of atmospheric salts and that there is a very strong decrease to the north where a minimum area seems to exist. It is, no doubt, the shielding effect of the mountains in South Norway which causes this minimum. Around the 64th parallel there is a broad pass in the mountains allowing a greater salt transport inland, very well indicated in fig. 1. Another interesting feature is that the spacing of the isolines seems to decrease considerably towards the Norwegian west coast. This is probably an elevation effect, the rise of the land from the sea being rather steep compared to the Swedish west coast.

As to Finland it is seen that the northern part is dependent upon the north Atlantic for its chloride while the southern part gets most of its chloride from the Baltic-Sea, though some may have come from more distant parts in southerly winds in which rain is known to contain considerable amounts of chloride (Rossby & Egnér l.c.).

A rather extensive investigation on the chemistry of river waters was carried out in the USA by the US Geological Survey a few years earlier than in Sweden, namely in the period 1905—1908. Water samples from 97 rivers east of the one hundredth meridian were taken frequently during a year's time and analysed for among others Ca^{2+}, Mg^{2+}, $Na^+ + K^+$, CO_3^{-2}, HCO_2^-, SO_4^{-2}, NO_3^-, Cl^- and total dissolved matter. A report of this investigation was published by Dole in 1909. In another report by Dole & Stabler the same year, yearly data on the runoff from a great number of rivers in the USA are given, together with computed yearly amounts of dissolved solids discharged by these rivers. The computations were to a great extent based on the chemical data published by Dole. From these it is possible to calculate the yearly amounts of Cl in runoff from 64 sampling points in the eastern half of the USA. These computed values have been plotted on the map in fig. 3 approximately in the centre of the drainage basins. For very large river systems greater weight has been given to the lower parts of the basins. Thus for the Mississippi River there are two values, 9.1 and 17.8 west and southwest of Lake Michigan, the first one representing samples collected at

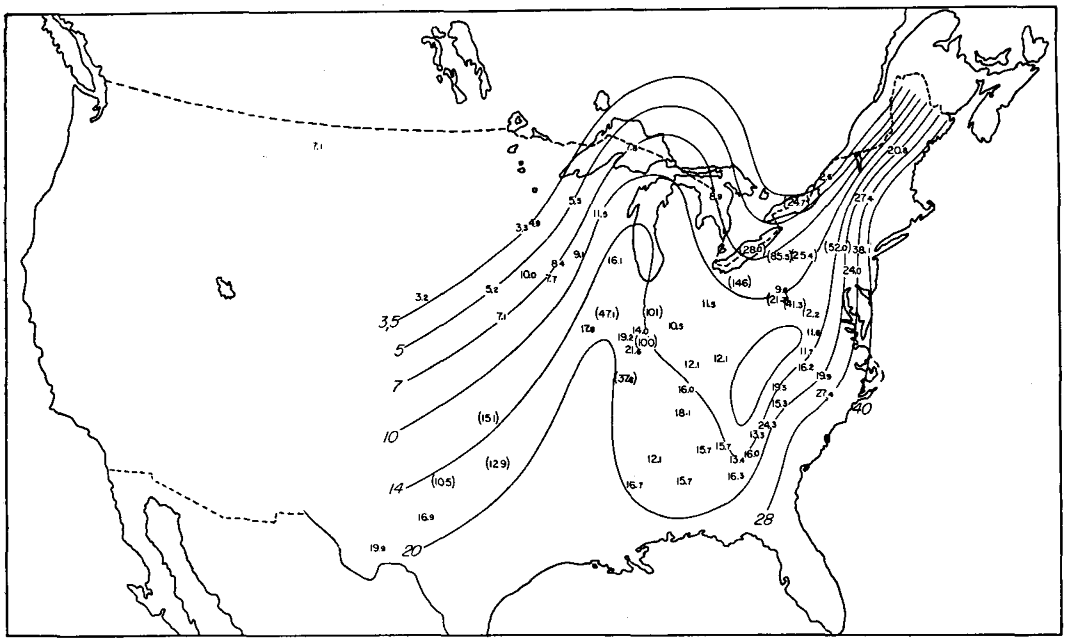

Fig. 3. Discharge of cloride by rivers in the eastern half of the United States in kg ha^{-1} year^{-1}. Isochlors logarithmically spaced.

Quincy, Ill., the second samples collected at Memphis, Tenn. The value 19.9 of the Rio Grande represents samples collected at Laredo, Texas.

On plotting the data on the map in fig. 3 it became evident that some values were of a different order of magnitude being up to ten times higher than the bulk of the values. On a closer inspection it appeared that these high values represented oil-producing districts like those of Oklahoma-Texas, Indiana-Illinois and Pennsylvania. It is well known that oil occurs together with salt domes; these high values must therefore be due to contribution from these salt domes. Values which can be suspected to be influenced by such salt deposits are put within brackets on the map and belong to the following rivers and sampling points.

	kg Cl ha^{-1} year^{-1}
1. Brazos R., Waco, Tex.	105
2. Arkansas R., Little Rock, Ark.	151
3. Red R. Shreveport, La.	129
4. Illinois R., Kampsville, Ill.	47.1
5. Big Muddy R., Murphysboro, Ill.	37.6
6. Embarras R., Lawrenceville, Ill.	100
7. Wabash R., Vincennes, Ind.	100
8. Muskingum R., Zanesville, Ohio	146
9. Allegheny R., Kittaning, Pa.	85.5
10. Monongahela R., Elisabeth, Pa.	21.7
11. Potomac R., Cumberland, Md.	41.3
12. Susquehanna R., Williamsport, Pa.	25.4
13. ,, ,, Danville, Pa.	52.0

It is obvious that Lake Erie receives some terrestrial Cl from such salt domes, hence the value 28.0 for this lake as well as the value 24.7 for Lake Ontario (St. Lawrence R.) have to be discarded. In this way a total of 15 values are neglected and the analysis of the map is based of the remaining 49. Of these one value, 14.0 from Embarras R. at Charlestone has been estimated by the author using about the same figure for total solids in runoff at that place as in the same river at Kampsville.

The isochlors are again logarithmically spaced, and reveal some interesting patterns. As it can be assumed that the Cl is mainly airborne it is seen that there are two main flows of presumably Cl-rich tropical maritime air from the south, one up through the Mississippi Valley and one up along the east

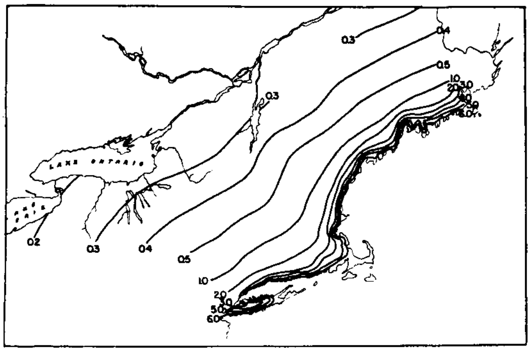

Fig. 4. Chloride concentrations in mg l^{-1} in natural unpolluted waters in New England and New York. (From Jackson 1905.)

coast, the well known track for Cape Hatteras storms and occasional tropical hurricanes. It is known that tropical maritime air from the Gulf of Mexico can reach farther north than the Canadian border. As there are no topographical obstacles in the broad Mississippi Valley the tropical air can reach very far north before precipitation removes its Cl content.

The low Cl values in the region between the Great Lakes and the east coast can be explained from the fact that most of the precipitation in this region comes from air masses from west which have passed over the mountain ranges in the west and lost its Cl on the western slopes of these mountains. This fact and the frequent passage of Cape Hatteras storm northeastward along the New England coast are responsible for the very steep gradient towards the east coast in this region. The same steep gradient is also seen on the maps of the normal distribution of chlorine in natural waters of New York and New England published by Jackson (1905). The normal chlorine map over this region published by him is reproduced in fig. 4 and it is seen that the pattern fits nicely into that of fig. 3. According to his isochlors the concentration near the coast amounts to about 6 mg l^{-1} while in the Lake Erie—Lake Ontario region it is only 0.2 to 0.3 mg l^{-1}. For an annual precipitation of 1,000 mm (40 inches) this would contribute 60 kg ha^{-1} $year^{-1}$ near the coast and 2 to 3 kg ha^{-1} $year^{-1}$ at Lake Erie—Lake Ontario.

Jackson's results have been discussed by Conway (1942) who used his map to calculate the total amount of sea salts brought annually over land from the sea by multiplying the amount of Cl per unit length of coast line in New England by the length of all coast lines. It is obvious that this will give too low an estimate.

There are very few data available on the content of chloride in precipitation in the USA. In a recent survey (1952) the author has listed three places, namely Geneva, N.Y., Mount Vernon, Iowa, and Madison, Kansas. At Geneva 18 kg ha^{-1} $year^{-1}$ was found as an average for the period 1919—1928 which compared to fig. 3 seems to be somewhat high. Collison & Mensching (1932) who presented this value admit that pollution by nearby sources was possible as there was a chemical laboratory in the neighbourhood. At Mount Vernon the estimate of Cl in precipitation is 72 kg ha^{-1} $year^{-1}$ which seems to be one order of magnitude too high. In Madison, Kansas, the estimate given is 18 kg ha^{-1} $year^{-1}$, which is in good agreement with the map in fig. 3. From these few data, however, it is not possible to discuss the role of possible direct transfer of Cl from condensation nuclei to the ground.

As to other parts of the North American continent there is very little information available. Rawson (1942) has published some data on lake waters from the eastern slopes of the Canadian Rockies west of Calgary. They are situated at altitudes between 345 and 1,663 m above sea level and with one exception (the southernmost, Waterton) show only traces of chloride. This can be expected as the air masses reaching these places would have lost their chloride in precipitation earlier.

It is possible from the map in fig. 1 to get some idea about the precipitation of other constituents like Na, K, Mg, Ca, H_3N, NO_3 and SO_4. However, their ratios to chloride in rainwater are not the same as in sea water; generally they are much higher in rainwater, as there may be other sources than the sea (probably distant ones) for these compounds. As to Ca this has been analysed for in the river water, and it may be interesting to compare the Ca/Cl ratio in the river waters with those in rainwaters calculated from the data given by Emanuelsson et al. (l. c.). In the river waters the Ca/Cl ratio varies from 0.4 to 1.5 with an average around 1 except for river water from limestone areas where the

Ca/Cl ratio may reach values up to 5. In the rainwaters the ratios vary rather systematically from about 1 near the west coast up to 3 or more in the eastern parts. From this it looks as if there were a net gain of Ca in most areas. As the excess Cl in river water compared to rainwater is probably due to the direct transfer of mostly giant condensation nuclei ($> 1\ \mu$) to the ground and as those are mainly composed of Na and Cl (JUNGE 1953) the Ca/Cl ratios in river water and rain water are not comparable. It is possible that the Ca in rainwater represents practically the total Ca delivered to the ground, though there are some indications in TAMM'S (l.c.) rainwater analyses that some Ca may be caught by spruce needles. In any case is seems safe to conclude that the denudation of Ca in soil and rock by percolating water is less than that inferred from river water analyses.

As to Na, it can be expected that the Cl/Na ratios in rainwater and river water should be comparable as both occurs in the particles $> 1\ \mu$. It can be mentioned that the Cl/Na ratio in lake waters in Sweden (calculated from the data of LOHAMMAR (1938)) is within the limit of those in rainwater. For Uppland (latitude 60° N, bordering toward the Baltic Sea) for instance, the ratios vary between 0.9 and 1.7 and if some lakes with very low electrolyte content are omitted there is a general trend in the ratios to increase towards the east coast. The same trend is also indicated in rainwater collected at different distances from the Baltic sea in more recent data, yet unpublished. Thus it looks as if Na in lake water is also derived to a very great extent from the atmosphere, and that it even can be relatively less in lake water than in rainwater, suggesting a net uptake by the soil (probably represented by moorland).

The composition of the precipitation when reaching the soil will apparently have a great influence on the composition of ground water as reflected in the river water. The original concentration will be increased not only by dissolved ions from the soil but very substantially by evaporation (including transpiration). As to Cl and probably Na the evaporation effect will be dominant. If the concentration of Cl in the precipitation, when reaching the ground, is C_N, the precipitation N and the evaporation E, then the concentration in the upper parts of the ground water will be

$$C = C_N \frac{N}{N-E} \tag{1}$$

It is seen that in a region where the evaporation is very near the precipitation very high concentrations can be reached. This may well happen in parts at the flat eastern central part of Sweden and is indicated in Lohammar's analyses from Uppland. GORHAM (1954) found in the analyses by Lohammar from Uppland and Dalecarlia an inverse relation between the total concentration of salts and the height above sea level. He attributed this relationship to the marine submergence of this region during glacial time and the progressive lifting afterwards, more sea salts being washed out in the higher regions than in the lower. As it happens there is also a general increase of precipitation with height above sea level which can be seen from Wallén's map (WALLÉN, 1951), and if the concentration of chloride is plotted against the amount of precipitation as estimated from his map a similar inverse relationship is obtained, showing the effect of evaporation, which at any rate is practically constant in this region compared to the precipitation.

RODHE (1949) made a very interesting study of Lohammar's lake water analyses. He found a remarkable similarity in the percentage composition of these lake waters, though the actual concentration differs considerably. This would be expected from eq. 1 if most of the salts in the lake waters were airborne, the concentration depending upon the $\frac{N}{N-E}$ ratio. This also has other implications, namely that the content of exchangeable ions in the upper parts of soils is to a large extent determined by two factors, the composition of the precipitation as it reaches the ground, and the composition of the organic matter which binds the exchangeable cations. As a steady state presumably has been reached the percentage composition of the precipitation will remain unchanged when percolating through these layers. Deeper down modifications may occur, especially with respect to Ca in our limestone areas, but additions of Cl is normally very unlikely and it looks as if Na too is mainly of atmospheric origin. As to the earlier submerged areas of eastern central Sweden it can be mentioned that MATTSON et al. (1950) investigated soil profiles on sedimentary clays and found only traces of Cl down to about 6 metres depth where a sudden increase occurred. This depth, however, was three metres down in the so-called soap clay, which is practically impermeable to water. As practically all the drainage occurs in the upper parts, the so-called dry crust, practically no chloride can come from the sedimentary clays.

It is apparent from the discussion above that the concentration of salts in ground water cannot be used as any measure of the chemical denudation neither qualitatively as suggested by many and most recently by Troedsson (1955) nor quantitatively as claimed by Arrhenius (1954). Arrhenius has presented a very interesting material, however, and revealed some very interesting relationships which ultimately may lead to a much better understanding of the hydrology of bedrocks but it looks from his data as if the deep groundwater contributes but little to the yearly water circulation. The river water is most probably derived from the uppermost parts of the ground water.

The best approach made so far towards the estimation of chemical denudation has been made by Viro (l.c.) who subtracted the salts in precipitation from those in river water.

Acknowledgment

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