Geochimica et Cosmochimica Acta, 1955, Vol. 7, pp. 129 to 150. Pergamon Press Ltd. London

On some factors affecting the chemical composition of Swedish fresh waters

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(Received December 1954)

ABSTRACT

An attempt is made to relate variations in the chemical composition of Swedish fresh waters (mainly analyzed by LOHAMMAR, 1938) to topographical, geochemical, and biological factors. The following are among the more general conclusions:

(1) In Uppland lake waters, and certain of those in Dalarna, total salt concentration is clearly related to altitude.

(2) There are distinct differences in ionic proportions between richer and poorer lake waters in both Dalarna and Uppland. In Dalarna the preponderance of moraine or Dalälven river sediments appears to be the important factor. In Uppland the high proportions of Cl, Na, and Mg in the waters of the lowest and richest lakes bear witness to the marine submergence of the district at the end of the glacial period.

(3) A comparison of Dalarna drainage and seepage lakes reveals that the waters of the latter are proportionally very rich in K but low in Ca. It is suggested that this may be due to a lesser degree of biotic influence on the waters reaching the seepage lakes.

(4) Dalarna seepage lakes exhibit low SO, levels in their waters, which may be connected with a slow rate of water transfer through them.

(5) The cations of bog waters are dominated by Na, those of fen waters by Ca. This illustrates the relative importance of atmospheric precipitation in the former, and mineral soil leaching in the latter. (6) The frequency distributions of dissolved plus particulate minor constituents in fresh waters

may be far from normal, low values predominating. Medians are therefore desirable in place of averages.

(7) Fe is most abundant in the more dilute waters, especially in Norrland. Mn is higher in the richer waters within each district, except in the Siljan area of limestone, where the lowest median occurs. Both elements tend to be highest (in respect of surface waters) in the shallower lakes.

(8) P is towest in the Siljan limestone lakes, and highest in the Dalarna seepage lakes.
(9) N is lowest in the most dilute waters, Norrland being particularly low, and higher in the richer waters within each district (except in the Siljan area).

(10) Sr, which like the major constituents is highly correlated with total ion concentration, forms a slightly larger part of the total ions in the more dilute waters, especially in Norrland.

INTRODUCTION

One of the largest and most complete bodies of data concerning the ionic composition of fresh waters is the treatise by LOHAMMAR (1938) on Wasserchemie und höhere Vegetation schwedischer Seen. LOHAMMAR's analyses have recently been examined in two interesting papers by RODHE (1949 and 1951), dealing separately with the major and minor constituents of the lake waters. This author has divided the lakes of the three Swedish districts investigated by LOHAMMAR into six groups, and calculated the correlation coefficients between the different ion concentrations and the specific conductivity of the water, which forms an estimate of the total salts in solution. The conclusion reached is that lake waters of the bicarbonate type exhibit a surprisingly constant composition as regards the major constituents, Ca, Mg, Na, K, Cl, SO_4 , and HCO_3 . This is believed to be the result of ionic exchanges between the dissolved ions and the colloidal systems of the soils and lake muds. RODHE then sets up the average ion proportions of the river and lake waters of the world (CLARKE, 1924) as a "standard composition" with which all fresh

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waters may be compared. For a range of total salt content from 15 to 350 mg/l, in the proportions of the "standard composition," curves are given relating individual ion concentration to specific conductivity. In this way a complete water analysis is not necessary for comparison, but only the concentration of the individual constituent and the specific conductivity of the water.

The total contents (dissolved plus particulate, in the unfiltered sample) of the minor constituents P, N, Fe, and Mn on the other hand, show little correlation with specific conductivity. This is believed to be due to (1) their largely un-ionized state, which is less affected by ion exchange, and (2) their low concentration, which is more affected by organism uptake, and in the case of Fe and Mn by reaction with the organic compounds in the water.

While RODHE recognizes that regional peculiarities may lead to differences in ionic composition (as evidenced in his Figs. 14–21, 1949), he does not discuss this aspect of the matter. The present object, therefore, is to draw attention to certain systematic deviations from the "standard composition" of fresh waters; and where possible to relate them to geochemical, biological, or topographical factors. Some features of interest with regard to the minor constituents, and to other fresh waters, are also discussed.

MAJOR CONSTITUENTS

In considering Ca, Mg, Na, K, Cl, SO_4 , and HCO_3 , RODHE deals chiefly with LOHAMMAR'S Uppland lakes, numbers 1–21. However, detailed examination of the Dalarna lakes is also of much interest. While the range of specific conductivity is less than in the Uppland series, the number of rather dilute waters is much greater, and certain points become apparent on this account. (It should perhaps be mentioned that, even so, the richer cultivated areas are over-represented in LOHAMMAR'S Dalarna material.) In addition, a number of seepage lakes lacking inflow and outflow streams are included among the Dalarna waters, and form an interesting contrast to the more abundant drainage lakes, which possess such streams.

Dalarna

Figs. 1–6 give the averages of LOHAMMAR'S analyses for the major constituents in the Dalarna waters, in relation to specific conductivity. All lakes from the Siljan area are marked as open triangles, whether seepage or drainage waters. The other seepage lakes are shown as crosses, and the other drainage lakes as dots. The curves inserted in the figures represent ionic concentration in relation to specific conductivity, for varying amounts of total salts in the proportions of the "standard composition."

If we look first at the drainage lakes outside the Siljan region, we see that up to a specific conductivity of about 75 the Ca values fall below the standard curve, while over 75 they are mostly above the line. In the Na figure the reverse is generally the case. It may also be remarked that none of the waters above a specific conductivity of 75 is (on the average) below pH 7, while several of the dilute waters are slightly acid.

We are here faced with a clear contrast. The dilute waters are often slightly acid, and are relatively rich in Na and poor in Ca. The more concentrated drainage lake waters tend to be alkaline, are rich in Ca and relatively poor in Na. A priori one might well seek the reason for these differences in the underlying geology of the areas draining into the lakes; and similar variations in the ionic proportions



Fig. 1. Calcium in relation to conductivity in the Dalarna lake waters.

of fresh waters have in fact been ascribed to the differences between the drainage from igneous and sedimentary rocks (CONWAY, 1942). PEARSALL (1921, 1922a and b) has also demonstrated differences in the ratio Na + K/Ca + Mg between



Fig. 2. Sodium in relation to conductivity.

rocky and silted lakes, lying partly in areas of volcanic rocks and partly in areas of sedimentary beds. The ratio is high in the rocky lakes, due chiefly to their low Ca levels. In the present instance, both the richer and poorer Dalarna lakes outside the Siljan district appear to be surrounded by the same granites and gneisses, as far as one can tell from the map series Aa of the Swedish Geological Survey.

Also to be considered is the effect of dilution upon the partition of mono- and



Fig. 3. Potassium in relation to conductivity.

divalent ions between adsorbed and solution phases of the soil-water system. MATTSON (1942) has shown that adsorbed monovalent ions should be strongly displaced into solution by divalent ions as the system is diluted, and the ratio Na + K/Ca + Mg is in fact higher in the more dilute lake waters. However, this is due solely to a great decrease in the percentage of Ca; Mg on the other hand is



Fig. 4. Magnesium in relation to conductivity.

about as much higher in the dilute drainage lakes as are both Na and K (see Table 1).

In this case the main reason is probably to be sought in the glacial history and topography of the region. The poorer lakes lie in predominantly morainic upland surroundings. Though some local sediments are present beside most of them, much of their drainage probably comes off glacial till. In contrast, the richer lakes lie mainly in the lowland sediments of the Dalälven river valley. (Two, numbers 58 and 77, do not. The first is influenced by nearby limestone, the second by lime-rich



Fig. 5. Chloride in relation to conductivity.

Cambro-Silurian morainic material.) This distinction, between lakes chiefly draining glacial till, and those draining Dalälven sediments, is reinforced by another factor dependent upon it. Cultivation and settlement are both much more extensive on the river deposits than on the till, and therefore agricultural liming, other fertilization, and pollution will be of much greater importance in drainage



Fig. 6. Sulphate in relation to conductivity.

from the former. Whichever of these two factors exerts the predominant effect on water composition, it appears reasonable to separate this lake group into two, those above and those below a specific conductivity of 75.

We may next compare the average cation proportions of the four lake groups in Dalarna, as they are given in Table 1. (It should be pointed out that the Siljan lakes are much the same, whether of the seepage or the drainage type; and the seepage lakes outside the Siljan area are likewise similar, whether above or below specific conductivity 75. Lake 81 has been excluded as abnormal, cf. LOHAMMAR, 1938. The following are the seepage lakes: numbers 26, 27, 29, 31, 46, 51, 59, 67, 79.) It is at once apparent that the poorer drainage lakes are proportionally higher than the richer ones in K and Mg as well as Na. However, the main feature

	Ca Ca +	$egin{array}{c} { m Na} \ per \ cent \ { m Na} + { m K} \end{array}$	K t of total + Mg by	Mg weight	Cl/SO ₄ ratio
River and lake waters, world average ¹	64	18	7	11	0.47
Uppland drainage lakes, 4–21 Uppland drainage lakes, 22, 24, and 25	69 51	16 29	4 4	11 16	$0.52 \\ 0.74$
Dalarna drainage lakes, conductivity < 75 Dalarna drainage lakes, conductivity > 75 Dalarna lakes, Siljan limestone area Dalarna seepage lakes	52 67 85 32	$23 \\ 16 \\ 8 \\ 24$	7 5 2 32	17 13 5 11	$0.57 \\ 0.49 \\ 1.0 \\ 4.1$
Ground water of a mid-Swedish podzol ²	40	37	5	17	
Finnish snow ³	14	48	20	17	1.5
Iron podzol, A horizon, mid-Sweden ⁴ Iron podzol, B horizon, mid-Sweden ⁴ Iron podzol, C horizon, mid-Sweden ⁴	26 25 24	28 24 27	40 39 37	6 12 12	
Average pre-Cambrian granite of Sweden ⁵	19	29	45	6	
Spruce forest ⁶ Pine forest ⁶	78 64	$2 \\ 5$	11 19	8 12	

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¹ Data compiled by CLARKE (1924).

² Data of TROEDSSON (1952), iron podzol on leptite-gneiss moraine. ³ Data of VIRO (1953).

⁴ Data of TAMM (1950).

⁵ Data of HOLMQUIST (1906), calculated by DALY (1914).

⁶ Data of EBERMAYER (1876), given by TROEDSSON (1952).

of the comparison is the extremely high K and low Ca of the seepage lakes outside the Siljan region. This latter district has on the other hand the most calcareous lake waters, with the lowest proportions of Na, K, and Mg. Since it is in fact an area of limestone, the proportion of Ca is not surprising.

A comparison of the Dalarna waters with the ground waters of a podzol, and with the composition of podzol soil materials and granites which predominate over much of the country, raises points of some interest. Data are given for these too in Table 1, calculated from the analyses of TROEDSSON (1952), TAMM (1950), and HOLMQUIST (1906). The interesting data of VIRO (1953) on Finnish snow are also included. We see that the composition of the seepage lake waters is intermediate between those of the granite and podzol materials and that of the drainage lake waters. These latter are much higher in Ca, somewhat higher in Mg, and very much lower in K than either the seepage lake waters, the podzol soils, or the granite. The podzol ground water resembles the drainage lake waters in respect of Mg and K, but is intermediate between seepage and drainage lakes with regard to Ca, and exceptionally rich in Na,

It is now desirable to attempt some explanation of these differences, even though our present knowledge restricts this to purely tentative speculation. Comparing their relative migration rates from the original igneous rocks to river waters, POLYNOV (1948) claims that on physico-chemical grounds of the solubility of their compounds, the order of migration of the major cations should be K > Na > Ca > Mg. This view is based partly upon calculations by the Russian geochemist FERSMAN (unfortunately no reference is given), and partly upon the order of the ionic potentials of the elements (i.e., the ratio of ionic charge to ionic radius, GOLDSCHMIDT, 1937; cf. also HUTCHINSON, 1943, and RANKAMA and SAHAMA, 1950). However, POLYNOV states that in fact Ca shows the greatest migratory capacity, and K the least (cf. also POLYNOV, 1937; CONWAY, 1942, 1945). If POLYNOV's method of determining actual migration rates is applied to the present waters, the order is Mg > Ca > Na > K, with the rates for Mg and Ca very similar. This is a complete reversal of the theoretical order given above. We may note that of the four elements. Mg is by far the least abundant in the Swedish rocks. REICHE (1942) has also noted deviation from POLYNOV's theoretical and actual orders in some New World areas.

POLYNOV's technique for calculating the relative migration rates of different elements or ions is to compare their proportional concentration in igneous rocks and in river waters draining igneous terrains, assuming that all elements in the latter are derived from the former. For example, from POLYNOV'S (1937) data, SO_4/Cl in igneous rocks = 3.0, in rivers from such rocks = 1.7; therefore mobility of SO₄ relative to $Cl = 1.7 \times 100/3.0 = 57$ per cent. While this method may be of some use with regard to the cations, a serious error in the basic premise occurs in the case of Cl and SO_4 , and vitiates at least partially POLYNOV's method of estimating migration rates. According to CONWAY (1942), almost all the Cl and SO_4 in river waters draining igneous terrains comes from sea-spray washed down in the rain. It is believed that the salts find their way into the atmosphere as droplets formed by the bursting of small bubbles (WOODCOCK et al., 1953; KIENTZLER et al., 1954) in foam patches formed either by high winds (WOODCOCK, 1953) or by the breaking of waves on beaches (BOYCE, 1951). Now, much of the Cl and SO_4 in the ocean cannot have derived from rock weathering (GOLDSCHMIDT, 1954; CONWAY, 1942), and may well have come from volcanic emanation (RAN-KAMA and SAHAMA, 1950). Therefore the above method of calculating mobility cannot be used for these two ions.

Even with cations, it is difficult to evaluate the contribution to fresh waters

from cyclic salts in rain, though it has been very crudely attempted (CONWAY, 1942; SVERDRUP, JOHNSON, and FLEMING, 1942; see also VIRO, 1953). While the cations may be presumed to have come originally from rock weathering, their return from the sea is unlikely to be in proportion to their delivery into it (cf. RANKAMA and SAHAMA, 1950). Nor can we expect them to be present in the proportions of sea-water, in view of the probable separation of salts in sea-spray during its evaporation. Japanese workers (MATSUI, 1944; MIYAKE, 1948; SUGAWARA et al., 1949; KOYAMA and SUGAWARA, 1953) have shown the SO_4/Cl and Ca/Na ratios of rain to increase greatly inland, and also with altitude of source. Their explanation is that evaporation of sea-spray separates crystalline $CaSO_4$, and to some extent MgSO₄, from hygroscopic NaCl particles. The latter can more easily form the nuclei of droplets, and presumably may be washed down in the rain more rapidly, while the latter may be carried like a fine dust farther inland. Dutch rain analyses also exhibit a rise in SO_4/Cl ratio inland, but some French data apparently show an opposite trend (see ERIKSSON, 1952). That rain and snow waters may vary widely in composition and concentration from one occasion to another has been demonstrated by various workers (TAMM, 1953; VIRO, 1953; GORHAM, unpublished). Lastly, it is necessary to separate the contribution of dust from that of sea-spray, which is extremely difficult even in non-industrial areas.

That sea-spray does provide an appreciable portion of the salts in rain seems likely from the above-quoted analyses. In this connexion, investigations of rain composition in progress at the Freshwater Biological Laboratory on Windermere have usually shown Na as the dominant cation, and the concentration of NaCl has been observed to rise sharply above the normal during westerly gales direct from the sea a few miles away. If sea-spray is important, its contribution tosurface waters should not be ignored in calculating migration rates. TAMM's (1953) figures for Ca, Na, and K in rain falling in the open (Table 2) show concentrations of much the same order as those in the less-rich Dalarna lake waters, the differences being least in respect of K and greatest in respect of Ca. Allowance for rain evaporation would bring the levels even closer. Also, in the English Lake District the total salt concentration of upland tarns may differ very little from that of many rainwater samples (unpublished work in this laboratory). Moreover, VIRO (1953) has calculated that for Finland as a whole, the material in precipitation is equal to about one-third of the total solids carried by rivers (including suspended matter). This proportion, admittedly based on a very few snow analyses, has also been calculated as 17 per cent for Ca, 30 per cent for Mg, 54 per cent for K, and perhaps about 99 per cent for Na. The SO_4 and Cl content of the snow, no doubt largely cyclic, appears to be more than equivalent to the sum of the four cations; and if NO_3 were added the anion excess would be even greater. The balance of cations is presumably made up chiefly of ammonium and hydrogen ions. The silica and sesquioxide contents of the snow are also quite appreciable, and probably derive from dust.

While the methods used to calculate migration rates have been shown to be extremely imperfect, it does seem likely that the major cations do not migrate in POLYNOV'S theoretical order (see also CONWAY, 1945), and he ascribes this to the intervention of organisms in the migratory process. Though of course discussion at the present stage must remain speculative, the situation with regard to the Swedish drainage and seepage lakes appears reasonably compatible with this view. The former, showing a much greater deviation from the ionic proportions of the granite, receive much water from streams draining surface-soil layers rich in both living and dead organic matter, and are much higher in Ca and Mg but lower in K than the seepage lakes. On the other hand the latter may be fed to a much greater extent by (1) little-altered rain as surface run-off (low in Ca and quite high in K —particularly if it has dripped through the forest canopy, see Table 2), and (2) slow-moving ground water long in contact with the deeper mineral soil horizons and rocks, and thus comparatively removed from biotic influence.

In this connexion it may be pointed out that the seepage lakes lie in deep pits along gravelly eskers, which form what may be described as subterranean streams. The gravels are usually of much the same mineralogical composition as the surrounding till, and their capacity to provide water is indicated by the fact that many Swedish towns pump their entire supply from such deposits (SJÖRS, private communication). Some drainage lakes occupy similar sites; but since their waters do not show differences from the normal ionic composition of drainage lake waters, its may be inferred that the surface stream drainage is of predominant importance. (It should, however, be mentioned that the highest K concentrations occur in seepage lakes most liable to agricultural contamination; it is not known how significant this factor may be.) LOHUIS, MELOCHE, and JUDAY (1938) have also found the residues of seepage lake waters in Wisconsin to be relatively rich in K, and to a lesser extent Na, as compared with the drainage lakes.

Although the seepage lake waters may be presumed to derive originally from surface precipitation, continued exchange during percolation through the deeper and wholly inorganic horizons of soil and rock might be expected to result in an alteration of their ionic proportions—toward those of a weathering medium unaffected by biological activity, in contact with the parent rock. RENICK (1924) has previously suggested cation exchange during percolation as the factor responsible for the chemical differences between deep and shallow well-waters in Montana. There shallow wells yield hard waters rich in Ca and Mg, while the deeper wells give relatively soft waters proportionally higher in monovalent ions; at the same time total salts are much the same in both. Similar phenomena have been reported by FOSTER (1950).

Of course it is not to be expected that the ionic proportions of even pure water in contact solely with the parent rock should be the same as those of the rock itself. Different minerals, and their different components, do not all dissolve with equal facility. Moreover, as regards ion exchange between rock decomposition products and water, BRAY (1942) has demonstrated that different ions are not all released from the adsorbed state with the same ease; and MATTSON (1942) has illustrated the effects of dilution upon such a system.

In view of all these factors, it is a matter of some interest that detailed investigations should be made concerning the sources, paths, and rates of water supply to the seepage lakes; and the composition of the neighbouring soils and rocks as well. Information regarding the cations adsorbed by the various soil horizons

would also be most valuable. Much of the argument in these pages rests upon assumptions which, though perhaps reasonable, urgently require verification.

The high levels of Na in the podzol ground water shown in Table 1, together with the low proportion of Ca as compared with the drainage lakes, may well be due to the high demand of the forest for Ca and its low requirement of Na. This too is exemplified in Table 1, from data of EBERMAYER (1876). TROEDSSON (1952) states that this ground water lies only one to two metres beneath the soil surface. so that a strong biotic effect seems likely. If the podzol ground water is in fact

Sample and location	Number of samples	Ca	Na mg per litre	К
Bain in open sites ¹	25	0.43	0.59	0.30
Rain beneath spruce and pine ¹	36	1.57	1.86	5.26
Ratio beneath trees/open sites		3.7	$3 \cdot 2$	17.5
Water from pools on raised bogs ²	9	0.29	0.80	0.23

Table	2.	Ionic	com	position	of	rain	and	bog	waters

¹ From analyses of TAMM (1953) in Uppland. ² From analyses of WITTING (1948) in Uppland.

drawn on to a greater extent by organisms than are the drainage lake waters, the result described above seems reasonable. It would also suggest that the dead residues of the biosphere are the most active agents in Ca liberation; presumably the humus acids (including biologically produced carbonic and sulphuric acids, cf. ROBINSON, 1949) could fulfil this role.

The marked reduction of K in the podzol ground water and in drainage lake waters, as compared with the seepage lake waters, snow water, podzol soils, and granite, is probably due largely to fixation of K in non-exchangeable form by colloidal clay minerals (cf. LEVINE and JOFFE, 1947). Fixation in sandy podzols and brown podzolic soils appears to be most active in the colloid-rich horizons (JOFFE and KOLODNY, no date); which may be said to have developed mainly as the result of biologically activated weathering and leaching (cf. KELLER and FREDERICKSON, 1952). Phosphate complexes of Fe, Al, Ca, and Mg also seem capable of fixing K (JOFFE and KOLODNY, 1937).

EBERMAYER'S (1876) data suggest that the K content of the forest is not very great, although many later workers claim a high demand for this element by plants. This contradiction may be largely due to differences in material or sampling. The analyses of HENRY (1908, cited by LUTZ and CHANDLER, 1946) point to a much greater Ca/K ratio in the stem, branches, and bark of trees than in leaves.

An interesting effect of the forest upon the cycle of K has been shown by TAMM (1953), and can be seen in Table 2. Percolation through the forest canopy can increase the salt content of rain about threefold in Na, fourfold in Ca, and as much as eighteenfold in K. How far this is due to leaching of the leaves, and how far to animal excrement, etc., is not known. In the open the rain is highest in Na, Ca is intermediate, and K is lowest-though by no means so low proportionally

as in the podzol ground water or the waters of the drainage lakes. Presumably a large part of the Na derives from sea-spray, and some of the Ca and K as well; but as mentioned earlier, the elucidation of dust versus sea-spray contributions is very difficult. After percolation through the forest canopy the ratio between the monovalent ions K and Na becomes much more like that in the living forest itself.

If we now turn our attention to the anions, the seepage lake waters are richer proportionally in Cl and poorer in SO_4 than those of the drainage lakes; and Cl



Fig. 7. The chloride and sulphate concentrations of Swedish river waters, in relation to total mineral content (data of ERIKSSON, 1929).

exceeds SO_4 in the former, while the reverse is the case in the latter. The waters of drainage lakes in Uppland, Norrland, and Skåne (ALMESTRAND, 1951) also generally contain more SO_4 than Cl. This seems difficult to reconcile with the data of ERIKSSON (1929), which indicate that the majority of Swedish rivers carry more Cl than SO_4 , especially in the north where soils are poor and both lake and river waters are extremely dilute. It is actually in these more dilute rivers that Cl predominates, in the most concentrated ones SO_4 is clearly in excess, as shown in Fig. 7. CONWAY (1942) has shown this to be a world-wide phenomenon. In Uppland SO_4 is well above Cl, while in Dalarna rivers it is only slightly less than Cl on the average, and yields a higher maximum value.

Swedish well-waters, which according to the data available are much more concentrated than river waters in the same area, also contain Cl in excess of SO_4 in most cases, but over the whole range of concentration (ARRHENIUS, 1952; since the data were provided by the public health service, waters suspected of contamination may be over-represented). The contrast between well and river waters is rather similar to that between seepage and drainage lakes, in that the

 Cl/SO_4 ratio is higher in the former in both cases. It is of interest in this connexion that RANKAMA and SAHAMA (1950) claim chlorides as predominant in salt solutions from cavities in granites, and in mine-waters, in spite of the fact that rocks usually contain more sulphur than chlorine. The Finnish snow (VIRO, 1953) also contains more Cl than SO_4 .

In the Dalarna drainage lakes, the more dilute waters exhibit a higher Cl/SO_4 ratio, as might be expected by analogy with the rivers.

A factor which has a marked effect upon the SO_4 regime of many lakes is the oxidation-reduction potential of the mud-water system. MORTIMER (1941-2) has demonstrated that through organic decomposition lake muds often become extremely reducing below; and that in shallow lakes during summer stratification, reducing conditions may even obtain at the mud surface, and in the bottom waters of the hypolimnion when oxygen is severely depleted. Under these conditions SO_4 may be reduced to sulphide, and if ferrous ions are also abundant and the pH is about neutral, black ferrous sulphide will precipitate. Dr. LOHAMMAR informs me that ferrous ions are sometimes plentiful in the bottom waters of the Dalarna seepage lakes during summer anaerobiosis, and hydrogen sulphide may be also detectable by smell.

If biological sedimentation of sulphur to the bottom, and its anaerobic precipitation in the muds, is not counter-balanced by inflow replacement, SO_4 may be expected to reach very low levels (BEAUCHAMP, 1952). This would be most likely in the seepage lakes, where water transfer may well be very slow. And those lakes with a rich plankton should be especially liable to SO_4 loss, in view of both the greater plankton sedimentation and the greater removal of oxygen through organic decay.

The Dalarna seepage lakes outside the Siljan area are in fact very low in SO_4 , with a mean value of 1.3 mg/l and several analyses below 0.5 mg/l. This may be compared with a mean of 5.8 mg/l in the Dalarna drainage lakes, which are only slightly higher on the average in respect of total salts. Moreover, LOHAMMAR (1938) has pointed out that SO_4 is particularly low in those seepage lakes with a large phytoplankton population. In this connexion BEAUCHAMP (1952) has suggested that SO_4 deficiency may limit the productivity of some tropical lakes, especially below 0.5 mg/l.

It should be mentioned that seepage lakes elsewhere do not exhibit the same SO_4 impoverishment. In Uppland, overlain by calcareous marl, and in the Siljan district of Silurian limestone, seepage lakes are only slightly lower in this ion than drainage lakes of similar specific conductivity. It may also be remembered that these seepage lakes are not very different from their drainage counterparts with respect to the major cations either. The writer can offer no explanation.

Another possibility to be considered is that if lakes without inflow streams do receive most of their water by slow diffuse seepage through the lake muds (and this is a point deserving investigation), SO_4 could be removed from circulation there by reduction and precipitation as ferrous sulphide. On the other hand, such precipitation would not seriously affect the epilimnion of drainage lakes obtaining most of their water from surface run-off and percolation through the upper layers of the soil, into well-aerated inflow streams. The SO_4 level of the summer-isolated

hypolimnion in drainage lakes can of course be lowered by reduction, as MORTIMER (1941-2) has shown.

The absence in May, July, and September data of any consistent summer fall of SO₄ in these Dalarna lakes needs study. Some much richer Uppland lakes with abundant rooted vegetation do exhibit a marked drop. With reference to this point, OHLE (1953) has suggested that fluctuations in ground-water level can strongly influence both the \overline{SO}_4 and HCO_3 status of fresh waters. These ions may show a reciprocal relation in ground water, SO_4 rising with a rising water table and HCO_{a} falling. The presumed explanation is that the sulphur compounds of soils (mainly sulphides of iron) may be oxidized to sulphuric acid and ferric hydroxide during periods of low water table when soil aeration is increased (see also WIKLANDER et al., 1949, 1950a and b). The sulphuric acid will then react with bicarbonate salts to yield sulphates, water, and carbon dioxide. Thus when the ground water rises again it will be enriched in SO_4 , which may wash out into the streams. Such a phenomenon might well have marked seasonal effects upon the inflow of SO_4 into lakes where the soils are rich in sulphur. WIKLANDER et al. (1949, 1950a and b) have shown that Swedish gyttja soils may in fact be rich in this element, occurring largely as sulphides of iron; and this is particularly true of the saline types. Thus a high sulphur content is a strong possibility in the areas of marine submergence in Uppland.

Many Norrland lakes are also low in SO_4 , the mean value (excluding two "sulphate lakes," cf. RODHE, 1949) is 1.9 mg/l. This low figure, however, reflects the general poverty of these waters in mineral matter: the proportion of SO_4 is only slightly below that of the "standard composition."

The correlation which often exists between a largely cyclic ion such as Cl and a mostly rock-derived ion like Ca is of some interest. Time may well be an important factor, allowing an increasing amount of material to be leached from the rock minerals into the percolating water, and also permitting evaporation, and consequently the concentration of all salts—including those thrown up in sea-spray and deposited on land by wind and rain.

Uppland

Some points of interest concerning the Uppland lakes may now be considered. Diagrams of ion concentration versus specific conductivity are given for lakes 1-21 by RODHE (1949). These show a wide range of total salts, specific conductivity varying from about 20 to 600; but on the whole the Uppland lakes are much richer than those of Dalarna, and particularly those of Norrland.

A most interesting feature of the total salt concentration is its marked dependence upon the height of the lake above sea-level, as illustrated in Fig. 8. The north-westerly lakes, being higher, possess less salt. The dilute Dalarna lakes on glacial till and with specific conductivity less than 75 exhibit a similar dependence (see Fig. 8), again with a slight north-westerly decline. The marine submergence of Uppland and parts of Dalarna at the end of the glacial period may be an important factor here. Deposits of layered clay are found over a large part of the region, and calcareous marl is abundant east of a line from Gävle to the tip of Mälaren (cf. Swedish Geological Survey maps Ba 2 and Ba 5; also LUNDQVIST, 1935). At the higher elevations the period of leaching, and presumably the leaching intensity (cf. ARRHENIUS, 1953) would be progressively greater. Another possibility is that in the uplands the amount of soil percolated by a given amount of rainwater on its way to a lake, and also the retention time within the drainage basin, may be less than in the lowlands.

PUKE (1949) has reported a similar decline of specific conductivity with altitude in the lakes around Stockholm. However, at heights much the same as those recorded by LOHAMMAR in Uppland, PUKE's specific conductivity values are rather lower, owing probably to a lesser degree of submarine marl deposition in his more



Fig. 8. The dependence of salt concentration upon height above sea-level, for all Uppland lakes and for Dalarna lakes on glacial till (i.e., below a conductivity of 75).

southerly localities. According to ARRHENIUS (1953), the mineral content of wellwaters in Dalarna also declines in a north-westerly direction as the mountains are approached. The above trend is, however, absent among the richer Dalarna lake waters. Evidently the lime content of the Dalälven sediments, and/or the degree of agricultural liming, is a more important variable in these sites.

Correlated with dilution, there is also a tendency toward acidity in the more elevated Uppland and Dalarna waters. LJUNGGREN (1953) has likewise shown a greater degree of acidity in Värmland lakes above the limit of marine transgression, with a tendency for the more acid waters to occur in the northern districts.

Of the four richest Uppland lakes, three are especially high in Na, Mg, and Cl, and low in Ca. Since they are all very near sea-level, this richness in the chlorides of Na and Mg (see Table 1) is probably a relic of their submergence, not yet obscured by leaching. The fourth lake, number 23, is located at a slightly higher elevation,

and according to LOHAMMAR (1938) may well owe its high salt content to excessive agricultural contamination.

It can be seen from Table 1 that the Uppland drainage lakes numbers 4-21 are very similar to their Dalarna counterparts, the drainage lakes above a specific conductivity of 75. Numbers 1-3 are below this value, and resemble the more dilute Dalarna waters. Lakes 3 and 10, although seepage lakes, do not show characteristics like those of the Dalarna seepage lakes.

Norrland

The Norrland lake waters are extremely dilute, and except for two "sulphate lakes" (cf. RODHE, 1949) are comparable with the Dalarna drainage lakes influenced chiefly by glacial till. They are therefore low in Ca relative to the "standard composition," and high in Na, Mg, and K. LOHAMMAR (1938) has noted a tendency to higher Na and Cl values nearer the coast, which may again be the result of a later emergence from the sea.

Swedish bogs and fens

Even more extreme deviations from the "standard composition" are observed in bog habitats. WITTING (1947, 1948) has determined the cation concentrations of waters from a series of bog and fen-plant communities, separated according to the

Plant community	Number of	Hp	Ca + Na + K + Mg	Ca	Na per cent	K of tote	Mg ıl	
· .	samples		mg/l	$\begin{array}{c} \textbf{Ca} + \textbf{Na} + \textbf{K} + \\ by \ weight \end{array}$				
Raised bog	43	3.9	3.3	16	58	11	15	
Fen window in bog	14	$4 \cdot 6$	4.0	27	53	6	14	
Extreme poor fen	15	4.3	4.4	38	45	5	12	
Poor fen	16	$5 \cdot 0$	6.4	28	46	7	20	
Fransitional poor fen	8	5.9	9.7	64	24	2	10	
Rich fen	4	$6 \cdot 1$	10.9	59	29	3	9	
Fransitional rich fen	5	$6 \cdot 5$	28.8	84	12	1	4	
'Hällkar'' poor fen near sea	11	4.4	34.0	8	70	4	19	
Extreme rich fen	7	7.5	53.4	81	10	1	8	
Sea-water		ca 8		3	84	3	10	

 Table 3. Cation proportions and acidity of bog and fen waters in Sweden,¹

 and in sea-water²

¹ Calculated from data of WITTING (1947, 1948).

² Calculated from data compiled by CLARKE (1924).

phyto-sociological doctrines of DU RIETZ (1949). The cation proportions and acidity of these are presented in Table 3, in order of increasing total content of the four cations. As a matter of interest the proportions of sea-water are included.

The bogs, which obtain their mineral supply solely by atmospheric precipitation, show extremely high proportions of Na. Moreover, in respect of the three cations

Na, Ca, and K, east Swedish bog waters are higher in the first and lower in the second and third than TAMM'S (1953) rain water samples from open sites in east Sweden (see Table 2). Ca and K may be taken up by plants to a greater extent than Na. Also, Na will presumably be adsorbed by the peat colloids to a lesser degree than Ca (MATTSON, 1942; BRAY, 1942). Why K should be reduced to nearly the same extent as Ca (when compared with the rain) is something of a mystery in these wholly organic and doubtless phosphate-deficient peats, but more analyses are clearly necessary. Some data of MATTSON and KARLSSON (1944) suggest that the uptake of K relative to Ca may be greater than is indicated by EBERMAYER'S (1876) data for forest (see also HUTCHINSON, 1943). While the above authors only analyzed the green parts and twigs of the bog plants, the woody parts (presumably high in Ca and low in K) are undoubtedly much less important in bogs than in forest.

A notable feature of Table 3 is the marked replacement of Na by Ca as the richer fens are reached and pH rises. The balance between atmospheric mineral supply and soil leaching appears here very clearly. K and Mg tend to be low throughout, but are highest proportionally in the more acid sites. The acid but salt-rich "hällkar" poor fens are situated in rock pools beside the sea and receive much spray directly, which explains their high Na/Ca ratio. In solutions made up chiefly of chlorides and sulphates of sodium, acidity can develop unchecked even at high salt concentrations, which indeed tend to displace the hydrogen ions produced by organic decay into solution from the adsorbed state. In contrast, where high concentrations of calcium bicarbonate are produced by leaching of the mineral soil, humus acids are effectively neutralized.

Differences in ionic proportions such as these are probably of great importance in conditioning the distribution of plants in wet habitats. Some of their possible effects in nature have been remarked by PRIESTLEY and HINCHLIFF (1922; HINCHLIFF and PRIESTLEY, 1924) and by PEARSALL (1922*a* and *b*; 1924). More recent studies have demonstrated the influence on plant growth (and presumably distribution) of varying ionic ratios in both culture solutions (OLSEN, 1935, 1942, 1950, 1953) and the adsorbed phase upon soil colloids (JENNY and AYERS, 1939; JOFFE and ZIMMERMAN, 1944; VLAMIS, 1949; HESLEP, 1951; EPSTEIN and STOUT, 1951).

MINOR CONSTITUENTS

If frequency distributions are constructed for LOHAMMAR'S analyses of dissolved plus particulate minor constituents, they appear very far from normal. Low values predominate, but there are occasional very high ones. Therefore it is perhaps better to compare median levels instead of averages for the various lake groups. Such a comparison is made in Table 4, where all determinations from each lake in the group have been included, without averaging within lakes. While this method is open to criticism, the number of analyses is insufficient to provide reliable lake medians, and the general median is perhaps the best alternative. The medians have been obtained by intersecting cumulative frequency distributions at the fifty per cent level; and are only very approximate since the curves were fitted rather roughly by eye. Similar medians are given for specific conductivity within the lake groups, as an indication of total salts. On some factors affecting the chemical composition of Swedish fresh waters

While Table 4 is not of course susceptible of complete explanation, certain points of interest may be remarked.

Lake group	$Specific \ conductivity \ mho \ imes \ 10^6, \ 20^\circ C$	Fe mg	Mn per litre	Ρ μι	N 9 per litre
Uppland drainage lakes, 4–21 Uppland drainage lakes, 22, 24, and 25	200 415	0.25 < 0.05	0·026 circa 0·035–0·040	41 41	300 high but extremely variable
Dalarna drainage lakes, conductivity < 75	45	0.18	0.020	20	240
Dalarna drainage lakes, conductivity > 75	90	< 0.05	0.033	20	340
Dalarna lakes, Siljan limestone dis- trict	115	< 0.05	0.008	12	220
Dalarna seepage lakes	55	< 0.05	0.020	45	230
Norrland drainage lakes, 101–165	25	0.83	0.019	20	110

 Table 4. Minor constituents (median values) in lake groups of

 Uppland, Dalarna, and Norrland

Iron

The Norrland lakes are clearly much higher than any others in this element. This is no doubt due to the general base deficiency of the northern rocks and soils, which has led through acid leaching to mobilization of iron from the soil minerals, and the marked development of podzol profiles (TAMM, 1950). The low total salt content of the lake waters reflects this situation. In Dalarna and Uppland (also in Skåne, cf. ALMESTRAND, 1951) the richer lakes tend to be lower in Fe, and the Siljan lakes are especially poor, with 31 of 33 analyses less than 0.05 mg/l. This last fact is presumably connected with the circum-neutrality of subsoils in this area. In spite of the usual poverty of limestone in Fe (RANKAMA and SAHAMA, 1950), the Siljan subsoils appear quite rich in this element (ARRHENIUS, 1953). It is not clear to the writer why the Uppland and Dalarna drainage lakes, differing widely in total salts, should have much the same levels of Fe and Mn, and show similar trends in relation to specific conductivity.

Not only are the poorer waters richer in Fe, there is also a greater deposition of lake and bog iron ore in poorer and more acid districts (LJUNGGREN, 1953). Thus the absolute difference between rich and poor areas with respect to iron leaching must be much greater than the differences in lake-water Fe suggest.

Outside Norrland it appears that only lakes less than five metres deep show Fe concentrations above 0.5 mg/l in the surface waters. EINSELE (1938) and MORTIMER (1941-2) have demonstrated that oxygen depletion during the summer may result in reduction of ferric complexes at the mud surface, leading to an escape

of divalent ferrous ions from the bottom, and a rise in Fe concentration in the lower levels of the water column. In the more shallow lakes this effect may penetrate to near the surface. JUDAY, BIRGE, and MELOCHE (1938) have similarly shown a marked increase of both Fe and Mn with depth in Wisconsin lakes, associated with a fall in oxygen and a rise in carbon dioxide. Turbulence may also result in stirring up precipitated ferric hydroxide from the mud surface to the upper levels of the water column in the shallower lakes.

The last-mentioned authors further state that Fe is higher in wells than in lakes from the same area. The Dalarna seepage lakes, which might perhaps be expected to show a similarity here, are low in Fe at the surface, possibly because better aerated. However, Dr. LOHAMMAR informs me that they are sometimes rich in Fe in the badly aerated deeper layers.

The effects of oxidation-reduction conditions upon iron may also be of importance in connexion with the concentrations of rarer elements in lake waters. GOLDSCHMIDT (1937, 1954) has suggested that in the sea many of the rare elements such as Se, As, Pb, Sb, and Bi are removed from solution by adsorption upon oxidized ferric colloidal complexes. He claims that in this way the possibly harmful biological effects of many poisonous substances are minimized. It would be of great interest to know how far this process operates in fresh waters, and whether summer reduction of ferric hydroxide and other complexes at the mud surface may have unsuspected effects upon fresh water organisms, at least in or near the mud. Since many trace elements may be beneficial in small amounts while toxic at higher concentrations, the effects might well be extremely variable. While positively charged ferric sols adsorb anions, negatively charged manganic sols can adsorb cations. For instance Mo, B, Co, Ni, Cu, Zn, Tl, Pb, W, etc., may concentrate in Mn sediments. Ferric organic complexes may of course have dual adsorptive powers (MATTSON, 1938; MORTIMER, 1949).

Strongly reducing conditions, under which Fe may precipitate as ferrous sulphide, may also bring about the precipitation of a number of rare elements, such as Pb, Cu, Zn, Mo, Tl, V, Co, Ni, etc. (GOLDSCHMIDT, 1954). This too may have important biological consequences in fresh waters.

Manganese

This element, like Fe, exhibits very low concentrations in the Siljan limestone lakes, though the subsoils and limestones are quite rich in Mn (ARRHENIUS, 1953). Again, the high pH of the subsoils may be responsible. However, Dr. LOHAMMAR informs me that lake number 80 is very rich in Mn in the deeper and probably badly aerated waters. Apart from the Siljan district, Mn tends broadly to show a trend opposite to Fe, being slightly lower in the poorer and higher in the richer lakes within the three geographic regions. Since the amount of Mn in granites is very much less than that of Fe (ARRHENIUS, 1953), the poorer soils and waters may be depleted of more soluble forms of this element. The data suggest that the compounds of Mn formed during weathering may be as a rule more stable in solution than those of Fe in lakes with a high salt content. There is some indication that the highest Mn values, like those of Fe, occur in shallow lakes.

With regard to bogs and fens, WITTING'S (1947, 1948) data show only low Mn

values (< 0.011 mg/l) in waters above pH 6.5. Greater amounts are most likely in the pH range 4-5 (median roughly about 0.025 mg/l), though five of the six highest individual figures come from fens between pH 5.5 and 6.5.

Phosphorus

Here again the Siljan lakes are low, in spite of a moderately good supply of citric acid-soluble P in the subsoils (ARRHENIUS, 1953). The presence of abundant subsoil calcium carbonate, and the associated high pH values, may well account for a low water solubility of phosphorus. The Dalarna seepage lakes are highest in total P, being more than twice as high as the drainage lakes in this area. This may be due both to a greater degree of direct biological removal from the waters reaching the latter, and to fixation (as in the case of K) in those horizons of the soil which are rich in colloids as a result of intensive weathering and leaching under biotic influence (HUTCHINSON, 1952; ROBINSON, 1949). Undersea precipitation of calcium phosphate (cf. RANKAMA and SAHAMA, 1950) during the marine submergence of Uppland may well have enriched the soils of the marl areas. This, plus a greater amount of agricultural contamination, and also a shorter period and lesser intensity of leaching, may be responsible for the rather high P levels of lakes in the province.

Nitrogen

The Norrland lakes contain the least N. This is probably a result of low organic production, which might be expected from the northerly location, and from the poverty of the rocks in soluble salts—reflected in the dilute character of the lake waters. The next poorest group is that of the Siljan region, though the lakes there are not significantly different in this respect from the other Dalarna lakes, except those on Dalälven sediments. While not poor in total salts, the deficiency of P in the Siljan waters, coupled with the low proportions of other cations in relation to Ca, may perhaps limit biological productivity. It is noteworthy that the N/P ratio is highest in the Siljan lakes, with the richer Dalarna waters running a close second. The higher figures for N tend to come from lakes richer in salts, though as RODHE (1949) has shown, the relation is extremely variable.

Strontium

RODHE (1951) has proved that Sr, though present in small quantities, is strongly correlated with specific conductivity in the three geographic regions. He suggests that this is so because of its occurrence on the ionized form, and its lack of participation in biotic metabolism (see, however, HUTCHINSON, 1943; ODUM, 1951). In addition, it may be remarked here that at lower levels of specific conductivity Sr accounts for a slightly larger part of the total salts, particularly in Norrland. The richest Uppland lake, number 25, has an exceptionally low percentage of Sr in its salts.

The ratio of Ca to Sr in these waters is also not without interest. It ranges from about 100 to 2,000, being lowest in the dilute waters, especially those of the north. With the exception of lake nember 165, this ratio does not exceed 450 in Norrland. The highest ratio comes from the most concentrated water, lake number 25 in Uppland.

Using the average Ca content of HOLMQUIST'S (1906) average Swedish granite, and LUNDEGÅRDH'S average Sr content of Swedish igneous rocks (RANKAMA and SAHAMA, 1950, Table 15.1), the Ca/Sr ratio works out at about 90. Therefore Sr appears to be notably impoverished relative to Ca in many waters, presumably owing to the lower solubility of its carbonates and sulphates. The Ca/Sr ratio of limestone (from Table 5.52 of RANKAMA and SAHAMA, 1950) is about 400–700, a range surpassed by many of the values recorded for the richer Swedish waters, including some from the Siljan limestone district.

From the data of WITTING (1947, 1948) it appears that a similar situation exists in bogs and fens. In the composite group "bogs to transitional poor fens" the median for the Ca/Sr ratio lies somewhere about 300; while in the "transitional rich fens" and "extreme rich fens" the ratio ranges from about 650 up to 2,550. The "hällkar" poor fens receiving direct sea-spray ranges between 60 and 160. According to GOLDSCHMIDT (1937) and RANKAMA and SAHAMA (1950), Sr is strongly enriched relative to Ca in sea-water (Ca/Sr ratio = 31). If, as ODUM (1951) claims, the Ca/Sr ratio in shells is about the same as that in the aquatic environment over a series of concentrations, the lowering of the ratio in the ocean must be non-biological, and due presumably to inorganic precipitation of Ca, which of course is much more abundant than Sr. The high ratios of the richest Uppland lakes, which still show traces of marine submergence in their high proportions of Na and Cl, are rather interesting in view of the low ratio in sea-water. The anomaly is probably due to re-solution of calcium carbonate of high Ca/Sr ratio, deposited during the submergence.

The "poor fens" of Aneboda seem also to have Sr enriched relative to Ca, with Ca/Sr ratios varying from about 50 to 120.

CONCLUDING REMARKS

It can be seen from the above discussion that much of the variation in chemical composition of Swedish fresh waters is susceptible of explanation in terms of topographical, geochemical, and biological factors. While it is of course impossible at present to answer with certainty many of the questions raised here, it is hoped that these speculations may at least serve to focus attention upon some of the problems posed by LOHAMMAR'S invaluable series of analyses.

Acknowledgments—It is a pleasure to express my warmest appreciation to Dr. GUNNAR LOHAMMAR, who from his detailed knowledge of the individual sites has kindly suggested many improvements in this treatment of his data. I should also like to thank the following persons for helpful criticisms of the MS: F. R. HAYES, G. E. HUTCHINSON, F. J. MACKERETH, C. H. MORTIMER W. RODHE, H. SJÖRS, C. O. TAMM, and T. TROEDSSON.

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