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**IMPORTANCE OF BIOGEOCHEMICAL PROCESSES AND
WEATHERING FOR NORWEGIAN STREAMWATER CHEMISTRY**

by

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IMPORTANCE OF BIOGEOCHEMICAL PROCESSES AND WEATHERING FOR NORWEGIAN
STREAMWATER CHEMISTRY

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Surface water acidification and fish decline in modern times has been taken as examples of the harmful effect of the sulphur emission from the modern world industry. Especially in Sweden and in Norway, several thousand lakes have lost their fish population in the 20th century, mostly after 1950. However, many lakes have been said "always" to have been barren, and these lakes have also proved to be acid (K.M. Strøm, 1925). In other cases we know of several lakes which lost their fish population already in the 19th century, (Helland, A., 1900-1903, Dahl, K., 1921, Torgersen, H., 1934). In these cases we know that the reason for the fish decline is the chemistry of the lake water, mainly the low pH and content of aluminum etc. In other parts of the world with no pollution or pyritic rocks, for instance in South American laterite areas with tropical podzols, very acid rivers at pH around 4, have been known for a long time (Sioli, H., 1977).

There seems to be general agreement that the modern industrialized society emits to the atmosphere around 70 million tons sulphur, corresponding to about 200 million tons of sulphuric acid per year, from combustion of fossil fuels and from metal smelters. This is about 1/3 of the total sulphur precipitating from the global atmosphere. 2/3 of the total sulphur precipitation thus have natural origins, partly from sulphate salts in sea spray, which during stormy weather is brought up in the air. To a greater extent, sulphur from rotting organic material, where the sulphur bearing amino acids in the proteins from the flora and the fauna in the oceans and on land, is passing through complicated chemical processes. Finally, they oxidize to sulphate and then fall back to the ocean and on land surface. The volcanos seem to contribute only a minor part of the sulphur emission to the atmosphere, that is, 2-4%.

It seems as if the Scandinavian problem of modern acidification of water courses, has been taken as a typical example for the harmful influence of atmospheric pollution. Because thousands of lakes in Southern Norway and Sweden have lost their fish population, this massive fish decline has been

taken as proof for acidification due to atmospheric pollution, ^{mainly} because it coincided in time with ^{an} ~~the~~ increased ⁱⁿ industry and the installation of high stacks which ^{further} spread the flue gases from ~~the~~ European industry after World War II.

It has been claimed that the acidity in ~~the~~ rain has increased 10 to 100 times, and that the acidity of surface water has followed this increase directly. Based on sulphur content in cores from ombrogenic peat, ~~N.A.~~ Sørensen (1983) ^s concludes that the sulphur content of the rain fall in Southern Scandinavia, has only increased by ^v about 70% from pre-industrial time, and not by a factor of 10, as claimed by some environmentalists. In addition to the lakes which have been scientifically reported to have lost their fish population in the 19th and 20th century, we have in our folklore, stories about streams and lakes which suddenly lost their fish population. This was then, in medieval time, taken as a divine punishment for sinful behavior of individuals, or the whole community.

In all those cases where lakes or streams have become barren in this century, we observe that the water episodically or permanently, has low pH values, mostly between 4 and 5. In most cases we have no reliable measurements of acidity prior to the fish decline. Investigations of bottom sediments from lakes may in some cases show quantitative changes in the diatome flora.

Although, there are many factors of importance for algae growth, the diatome composition has hypothetical been taken as ^a indicator of acidity. According to these investigations, ^{there} ~~it~~ seems ^s to have been a decrease of less than ^e one pH unit in modern time - (corresponding to the upper 10 cm of the lake sediments). In an undated, probably less than 2000 years old sediment core, Berge (1975) however, ^{claimed} ~~reported~~ to have found greater variations deeper down. According to the formulae used, he calculated ^a the pH value of less than 5 at 20 cm depth and less ^{than} 4.3 at 78 cm depth in the sediment. In our institute, Dr. Bjørg Stabell, have also recorded considerable variations in the diatom ^{with depth} ~~up and down~~ in some sediment cores. This is following variations in the pollen examined by Dr. Helge Høeg.

The acidity of the rain fall in the investigated areas, of southern Norway and Sweden, is relatively low and fairly homogeneous, corresponding to around 40 micro equivalents H^+ per liter ~~50%~~ ^{an} on annual basis. (This again is varying by 50%)

correspond^s to one small glass of fresh lemon juice dissolved in one ton of distilled water). On the other hand, H^+ in streamlets in gneiss-granitic areas, varies within factors ^{of} ~~of~~ 1:1000. (If gabbro areas are included we have variations ^{of} ~~of~~ 1:10000). We know today that there is no direct connection between acid episodes in streamwater and acidity in the rain^e fall prior to measurements of the streamwater pH.

We know, however, that acid episodes in the streams always occur when the streams are in flood following heavy rain storms or snow melts, and that the most acid rivers are found in areas with the highest precipitation. In all cases, episodic rain storms of high or low pH, or low or high sulphate contents, are followed by episodes with low pH in the river (Skartveit et al., 1980). The pH, however, varies from streamlet to streamlet, on the same type of bed-rock.

Frequently, run-off water may be much more acid than the rain, and in small creeks within a very limited area, with the same rain and the same bed-rock, we may have very strong variations in the acidity in the run-off water. At base-flow, all these rivers have low acidity, often only 1/20 of the H^+ content of the same streamlet at storm flow. The most important factors today seem to be the hydraulic run-off pattern and ion exchange processes in the top soil, combined with weathering processes in the deeper parts of the profiles.

All acid streams and lakes in Norway, are found in areas with acid podzolic soil profiles. Frequently, we have off-shortened podzols containing only the mor, O layer and the A layer. There are today sufficient data to say that a given amount of precipitation will give the same acidity, or H^+ concentration, in the run-off water from a given catchment, provided the ionic strength in the precipitation water is the same. Variations in the sulphate/chloride ratio does not lead to any noticeable differences.

By laboratory experiments it is shown that equivalent diluted sulphuric acid, calcium sulphate solution, or calcium chloride solution, give^s exactly the same pH value of water ^{after it} ~~which~~ has percolated through the top soil from the water-shed of an acid river. The acidity will, however, vary from field to field, depending on two factors - the run-off hydrology and the nature of the soil profiles.

Changes in vegetations^g, and consequently changes in accumulation or ← oxydation of components in the biogeochemical environments, are dominating factors. The organic material may occur as raw humus (mor) or mull. Mor will be active as pH-buffer at low pH values. Whereas, mull has much lower capacity and the pH of the soil water will give run-off water at high pH.

After the uncontrolled, unfenced goat- and sheep grazing was forbidden in the first part of the 19th century in Denmark, the sandy west Danish calluna heath areas (without carbonates in the underground) ha^ve^e undergone a transformation from mor to mull (since P.H. Møller examined those profiles in 1879 and 1884). Now these areas have been planted with leafwood - ash, elm and maple. The top soil has totally changed. Fleming Juncker (1985)^g estimated that the pH has increased by more than 1 pH unit, in spite of the fact that the rain is considerably more polluted than in central Scandina-
navia.

In Norway and Sweden, however, we have had a change in the vegetation in the opposite direction. Coniferous forests, spruce and pine and heather ← has taken over more or less naked land with low humus content, just in the areas where we today have the acidified rivers. (These areas were investigated in 1854 by the Prussian chief forest inspector, K. von Berg, 1855).
They were reexamined
~~This is now described~~ by Tryggve Troedesson (1984). The changes have led to ← formation of thick layers of acid mor during these 130 years. In Sweden, Per A. Melkerud^g (1985) investigated an area which was grazing land, ← at least from the beginning of the 17th century, but from 1820 the area has gradually been reforested by spruce.

It appear^s well documented that we (in this case) have had^v a strong chemical weathering and acidification after the 1st, 2nd and 3rd generation of spruce trees. This is followed by neo-formations of smectite minerals from the early mica and feldspar-sandy soil.

In podzol profiles in Sweden and Norway, Gunnar Jacks (1985) and I, have independently found there is a reservoir of exchangeable H^+ in the acid soil profiles, corresponding sometimes to many hundreds, or even^a thousand years of the present polluted rain-fall. This H^+ will, by ion exchange with electro positive ions in the water, be liberated^g provided there are ← sufficient anions. In all these acid areas, the profiles are out of equilibrium, so that the amount of organic material formed from the litter

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from the trees is greater than the oxidation. Whereas, in the natural forests, before the influence of man (some 4000 years ago), there must have been an equilibrium - so that the oxidation of organic material corresponded to the neo-formation.

As mentioned, we have no acid brooks, except in areas where we have acid top soil. On the other hand, there are areas with acid top soil and deep sandy feldspar-bearing sub-soils. In these cases we have no acid run-off.

During the International Biological Program, the amount of annual growth of sphagnum mosses was found to be between 100 and 300 gram dry material per m^2 per year in the sub-alpine parts of Norway. In these mosses the dry material contained polyuronic-acid-pentosane-aggregates corresponding to, from 18% to 30% polyuronic acid. That means we have a neo production of 18 to 105 gram per m^2 per year. As the equivalent weight of polyuronic acid is 176 gram, this means we have a neo-formation of between 100 and 600 milli-equivalents of exchangeable H^+ per m^2 per year. This may be compared to a 1000 millimetre rain^cfall with a pH of 4.3, which correspond^s to 50 milli eqv. H^+ per year. ←

Most of the newformed polyuronic acid cannot contribute to the acidification in surface water, as there is not sufficient amounts of anions in the rain. The material is then either oxydized and return^{ed}~~ing~~ to the atmosphere as carbon dioxyde and water, or it may form peat layers. ←

In some peat moors the amount of exchangeable proton^es correspond^s to several kilometers of the present polluted rain^cfall. The importance of the exchange processes, as the determining factor of the run-off water, is beautifully illustrated by Richard Wright and ^{his} ~~the~~ co-workers, in the so called project RAIN. (1983). In this case two mini-catchment areas in Risdalsheia, in Southern Norway, were covered by transparent roofs in the spring^{of} 1984. The water from one of these roofs was first de-ionized and ocean water was then added in order to give the same level of chlorine, as in the other field. This means about 80 micro equivalents chlorine per litre. Then, the treated low sulphate high pH water was spread on the catchment area under the roof. The other mini-catchment was also covered by a transparent roof, but the rain water was directly spread under the roof. ←

In the first case the artificial rain water had pH 5.05 and 7 micro equivalent sulphate per litre. Whereas, the rain in the field had water with pH 4.3 and 64 micro equivalents sulphate per litre. The total run-off in the first case, for the period June 13th 1984 to November 14th 1985, had pH 4.07 whereas, in the case where the untreated precipitation was spread under the roof, the pH was 4.03. In both cases the run-off water was more acid than the applied "rain". Difference between the two fields was, however, insignificant.

We have further to be aware of the fact that under the first roof, nitrate and ammonium had now been removed and we will probably, in a few years, observe a change in the flora in the field with the de-ionized water. This may increase the pH.

If we pass along a sloping field during heavy rain fall and take samples of the water which is running off on the surface, we frequently observe that the H^+ content varies within ^a factor 4, or more over a distance of 100 metre, and the run-off water may be 10 times more acid than the rain water. This shows that the ion exchange processes between the cations in the rain and the cations (mainly H^+) in the mor, is a very quick process, which always leads to acid run-off. Deeper down in the profiles, the weathering of the feldspar and other minerals, will again neutralize the acid formed in the upper layer. This is the reason for the much higher pH in the water courses ^{at} base flow.

The acidity of the run-off water from the top soil depends mainly on ^{the} nature of the mor and the content of mobile anions ^{such} as sulphate, chloride or nitrates. For each small area, there exists ^{at} high intensity of run-off, a linear relationship $H^+ = A \sum (Cl^-, NO_3^-, SO_3^{--}) + B$. (A and B vary from area to area).

In a soil profile the O-layer and the bleached A-layer, are nearly pure cation exchange material. Whereas, the B-layer may at the same time be cation and anion exchange.

In the acid soil profiles, the cation exchange layers, are often more than 90% in the proton state - and the ion exchange capacity is of the order ^{of} one gram equivalent per kilogram.

Also, the precipitation layers B are chiefly cation exchanger, but especially polarizable anions, like sulphate and phosphate, may here partly exchange on the (OH) of hydroxides and clay minerals and be retained.

A. Henriksen (1980)⁷⁹ presented correlations between alkalinity (mainly HCO_3^-) and SO_4^{--} and bivalent cations (Ca + Mg) in lake water. He gave the following statements: "The acidification of fresh water is a continuous process analogous to the titration of a bicarbonate solution with sulphuric acid" further, "A consistent characteristic of acidified waters is the replacement of bicarbonate by sulphate while little, if any, change in the relative concentrations of cations has occurred".

That these statements are wrong is obvious to anyone who has worked with ion exchange processes and hydrology in the field.

He could, as we have done, take one litre of the Oslo drinking water from the tap in his office, and 10 grams of the raw humus from the reforested spruce area outside the building - then he could rinse the mor material with deionized water, to get rid of all water soluble acids and subsequently filter the tap water through the soil. The analysis will give the following results:

Tap water

pH 6.4

$\text{H}^+ = 0.4$ $\mu\text{.eqv./litre}$

$\text{Ca}^{++} = 145$ " "

$\text{Mg}^{++} = 46$ " "

$\text{SO}_4^{--} = 150$ " "

$\text{HCO}_3^- = 43$ " "

Filtered through mor

pH 4.26

$\text{H}^+ = 55$ $\mu\text{.eqv./litre}$ M

$\text{Ca}^{++} = 25$ " "

$\text{Mg}^{++} = 10$ " "

$\text{SO}_4^{--} = \text{n.d.}$

$\text{HCO}_3^- = 0$ " "

N.A. Sørensen (1985) was probably the first Norwegian scientist who stressed the fact that the rivers and lakes where salmonide fish had high mortality were characterized by very low Ca content. This does not necessary depend upon extremely low Ca content in the precipitation or the bed-rock and rhegolite, but upon the fact that Ca is fixed in a biogeochemical ion exchange material which is out of equilibrium with the chemical environment in which it is situated. This is illustrated by the above listed change in chemistry due to the actions of the fresh, raw humus.

The importance of the organic layer is demonstrated in three examined places in South Norway (Rosenqvist, 1985). In these three localities, extensive forest fires in the dry season had burned off the total organic layer, 6 to 20 years before the examination. In all three cases the run-off from these areas had acidities 1000 to 4 times less than in the areas just outside the burned fields. On the other hand, the content of magnesium and calcium was found to be considerably higher in the run-off from the area which had burned 6 - 10 years before the investigations as compared to the run-off outside the area. This difference also existed, but to a minor degree in the two re-vegetated areas, which burned 20 years before. This has led some people to say that the bed-rock must have been richer in calcium and magnesium in the burned area than outside - and perhaps the waters always had had a higher pH ⁱⁿ of the burned area. Beside, the low probability that forests are more inflameable at high pH than ~~at~~ low pH we have investigated the effect of ion exchange on such waters. Kjell Hegna (1986) has from 1982 to 1985 examined the limnology of an area which burned ~~down~~ ⁱⁿ 1976 and compared it with a neighbouring area which did not burn, and had very acid water.

In our laboratory my colleague, Turid Winje, carried out the following experiment:

To an artificial solution of chlorides, sulphates and bicarbonates of Mg Ca Na K, corresponding to the neutral run-off from the field which burned 10 years ago, was added 0.1 - 0.05% humus (mor) from outside the burned area and the filtrate was also analysed

Original sol. micro eqv.

Filtrate micro eqv.

| | | | | | |
|----------------------|--------|-------------------------------|--------|------------------|-------|
| | pH 6.1 | | | pH 4.5 | |
| (+) Mg ⁺⁺ | 20.6 | SO ₄ ⁻⁻ | 70 | Mg ⁺⁺ | 3.5 |
| Ca ⁺⁺ | 85 | Cl ⁻ | 50 | Ca ⁺⁺ | 8 |
| Na ⁺ | 27 | HCO ₃ ⁻ | 28 | Na ⁺ | 53 |
| K ⁺ | 11.8 | | | K ⁺ | 25 |
| H ⁺ | 1 | | | H ⁺ | 30 |
| | | | | | |
| | + 14.4 | | - 14.8 | | |
| | | | | + 120 | - 123 |

We thus can see that the ion exchange, mainly of the Ca and Mg ions,

liberated univalent ions Na^+ , K^+ and H^+ . The H^+ first reacted with the bicarbonate ion and gave at the lower pH, CO_2 and water. After having neutralized the bicarbonate, further ion exchange lowered the pH. By increased contact with one gram humus per litre - Mg dropped ^d to 3, Ca to 6.5, Na to 50 and K increased to 27.5 - and pH dropped further.

The average composition of the quartz-bearing magmatic and metamorphic areas in Southern Norway is close to:

| | |
|-------------------------|-------|
| SiO_2 | 70% |
| TiO_2 | 0.5% |
| Al_2O_3 | 14.5% |
| Fe_2O_3 | 1.5% |
| FeO | 2% |
| MnO | 0.1% |
| MgO | 0.8% |
| CaO | 2.0% |
| Na_2O | 3.5% |
| K_2O | 4.0% |

The relative high percentage of Mg and Ca in the weathering rock is generally not found in the acid run-off water. In spite of the fact that plagioclase, biotite and amphibole are more attacked ^K than the other silicate minerals.

This clearly demonstrates ^s the fact that the bivalent ions have the highest capacity to exchange and liberate the H^+ from the ion exchange material. Thus, it is the fixing of Ca^{++} in the catchment which is the reason for the low Ca content in the run-off, and at the same time the reason for the high H^+ content.

The S and Cl in Southern Norway seem to be higher than ^{the} continental crust average, particularly ^y in the rocks of so called Bamble Formation. The neutralization capacity of rock and regolite, by weathering, may be calculated. Different methods have been used.

From the precipitated CBD (citrate, bicarbonate, dithionite) soluble part of the B-layer in one postglacial podzol weathered till ^R Rosenqvist, Jørgensen and Rueslåtten (1980) ^f calculated a minimum value ^{of} consumed H^+ of 140 micro eqv. per m^2 per year.

10

In Cascade Mountain, U.S.A., Reynolds and Johnson (1972) calculated 930 m.eqv. H^+ per m^2 per year.

The enrichment of zircon in the tills of upper Numedal in Southern Norway, compared to the zircon content in the precambrian rock, from which the till originated, indicates that half the silt fraction must have gone into solution (Stiberg, 1983). If this had taken place in 10 000 years (post Weichsel), more than 300 m.eqv. H^+ per m^2 must have reacted by weathering per year.

If we compare these figures with the H^+ content of 1000 mm rain^{of} fall, pH 4.3 = 40 m.eqv. per m^2 per year, we understand that if it had not been for the "rejuvenation" of the acidity due to the increasing raw humus (mor) by exchanging cations as Mg^{++} and Ca^{++} "from the weathering water" against H^+ , no acid surface water would exist in these areas by the relatively low degree of pollution. In fact, nobody has ever been able to point to any area in Scandinavia where the H^+ of acid rain makes up as much as 15% of the total H^+ budget. Even in the areas with the most acid run-off. Also, in areas with little regolite cover, the weathering on the barren granitic rock, has been of the same magnitude^{of the l.c.} — order of magnitude 3 - 10 μm per year, that is, 10 - 30 gram per m^2 per year since the ice-age.

Although, the emission of sulphur in United Kingdom (UK) was more than 50% higher in 1972 than today (UK Department of Environment, 1984) and the other western countries, including Scandinavia, have reduced their emissions in similar degrees - the number of acid rivers is steadily increasing in Norway.

By intense local emitters, as the Ni smelter in Sudbury (Dillon et al., 1977) we have very much higher acidity in the precipitation, and dry deposition, than in Scandinavia, but the lakes are rarely as acid as the Scandinavian barren lakes. We had from 1880 to 1946, an important Ni smelter in Norway at Evje. In a narrow valley this smelter emitted every winter more sulphate than is now falling over the surrounding 10 000 km^2 , but the lakes in the district had plenty of fish. Some 15 years after the smelter was closed down, some of the surrounding small lakes became barren together with the general fish decline in Southern Scandinavia.

One of these lakes is now being thoroughly^{g^h} examined by many methods, thanks to the British, Swedish, Norwegian SWAP program.

The hydrology of a catchment area in the acid areas may schematically be illustrated by a model consisting of two types of material: A) Ion exchange materials, mainly in the H^+ state, B) Rockforming minerals.

On its way to the water courses, rain water will pass both types of material. Depending upon variations in permeability and intensity of precipitation, water may first pass through material A) then through material B) and again be pressed up in material A) and so on. ^{During} ~~By~~ rain storms the influence of material A) may dominate. ^{e Az} ~~By~~ base flow material B) will be the dominating material factor. If the material of B) is of gneiss-granitic composition, as in the Scandinavian podzols, weathering processes will tend to neutralize the percolating water, but this will again turn acid by the passage through and upon material A).

By tropical podzols material, as represented in areas with acid rivers (Sioli) ¹⁹⁷⁷ ~~point~~ B) is latheritic and not capable of neutralizing acids.

If material A) is in equilibrium or absent, because it has burned off, material B) will dominate and the run-off will have high pH. Only if material A) is increasing (out of equilibrium) as in the present acid ^s ~~Scandinavian~~ ^{areas of} ~~areas~~, any rain ^{will} result in acid run-off.

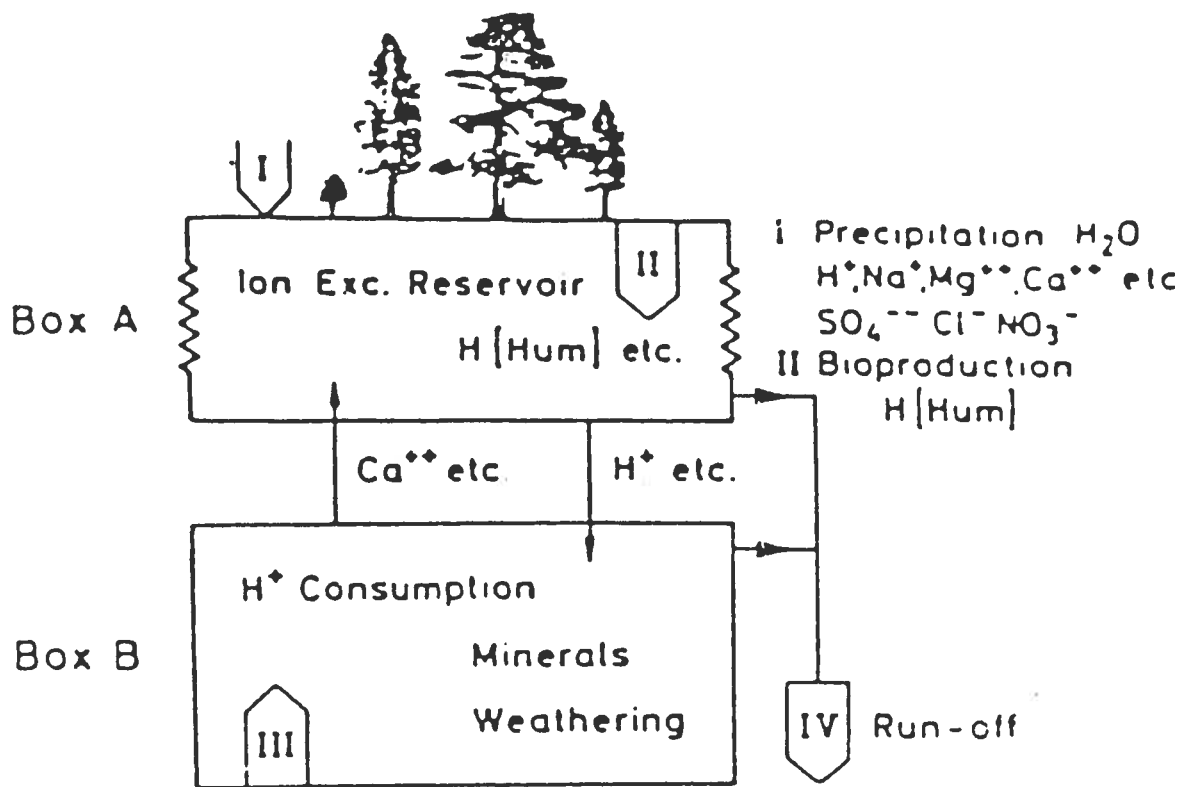
Fig 1

Closure

If you ask the right question, Nature will give you the correct answer.

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Type reactions



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