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Article abstract

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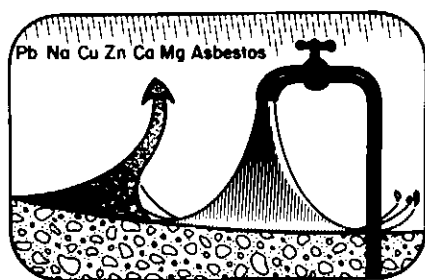
Quaternary Geology and Environmental Health

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Summary
This discussion considers two aspects of environmental health and Quaternary geology. One aspect considers the inhalation of dusts derived from unconsolidated sediments, and the other aspect considers ingestion and the "water factor". Both of these aspects of environmental health require mineralogical and geochemical information regarding Quaternary sediments, and most often there are little data available.

Introduction
Western man has greatly altered the surface of the earth by degradation of rock material and by removal of vegetation. The former mechanism, with a rate approaching the rate of natural processes has created fresh surfaces of fine-sized material, and a certain portion of this material can enter the respiratory system. Furthermore the increased surface area and the fresh mineral sufaces make the material more chemically reactive (Holdren and Berner, 1979; Berner and Holdren, 1979). The decrease in the vegetative cover results in an increased atmospheric particulate concentration, because there are fewer trees for fine particle interception and there is less ground cover to retain fine particles. The result is that in the last century chemical denudation should be increasing along with higher particle concentrations in the lower troposphere. Furthermore there should be increased atmospheric transport of fine particles.

The source of most of the fine particulates is unconsolidated sediments. Knowledge of their composition and mineralogy is needed in order to assess potential health hazards.

The following summarizes information regarding inhalation of mineral particles and ingestion of different types of water relative to disease and relative to Eastern Canada. Additional background information is found in Levander and Cheng (1980) and the Subcommittee on the Geochemical Environment in Relation to Health and Disease (1974, 1977, 1978).

History and Perspective
Most studies on environmental health and geology have emphasized water factors; trace metals have been the focus for solids. These studies have been carried out predominantly in the past 10 to 20 years. Kobayashi (1957) was probably the first to note a correlation to water quality for a non-occupational population; he found that cerebrovascular accidents increased with high sulphate: carbonate river waters in Japan. Schroeder (1960a, 1960b), recognizing Kobayashi's work, showed an association between deaths from hypertension and arterisclerotic heart disease and increasing soft water for the U.S.A. This association, for large populations, has been shown to exist for most of the World, for example, in Great Britain (Morris, *et al.*, 1962), Sweden (Biörck *et al.*, 1965), Canada (Neri and Day, 1968), and Finland (Karrpanen *et al.*, 1973). The World Health Organization has a series of ongoing projects related to geochemical factors and health, focusing especially upon the inverse relationship of cardiovascular disease and water hardness (Masironi, 1977). Marier *et al.* (1979) have reviewed the water hardness factor for Canada; they emphasize the role of magnesium only, however, and ignore other associated factors.

The literature on inhalation of soil dust and disease is quite sparse except for occupational setting such as with asbestos. And with the exception of fibrous minerals, most of the information relates to the trace metal geochemistry instead of including the mineralogy. In a search of the literature, the works of Burlikov and Michailova (1972), Burlikov *et al.*, (1972) and Mikhailova-docheva (1972) are apparently the only studies of endemic respiratory disease and soil mineralogy for a non-occupational setting. This is somewhat surprising in that it has been known for 10 to 20 years that there is a wide variety of fibrous and non-fibrous minerals and non-minerals found in the lung in non-occupational and occupational settings (e.g., Cralley, 1970). There have been laboratory animal injection studies on effects of various minerals, however (e.g., Hunter and

Thomson, 1973; Koshi *et al.*, 1968; Pott *et al.*, 1974; Pylev *et al*, 1975).

A new dimension on respiratory disease and geochemistry is suggested by the recent projections of Hamilton (1979) of excess deaths due to acid sulphates which have a greater irritancy than sulphur dioxide. The acidity of these particles will certainly be affected to some degree by the interaction with ambient atmospheric dust. Most of this dust will reflect the regional mineralogy and geochemistry.

Straf (1980), commenting upon the British Regional Heart Study, states that the inhalation pathway and the ingestion pathway should be studied together in epidemiological programs. This is most significant in that both the aerosol and water characteristics are due to a great degree to the mineralogical and geochemical characteristics of the unconsolidated elements.

Inhalation
Consideration of particles in inhalation divide practically into fibrous and non-fibrous minerals. Asbestos minerals have been studied in quite some depth due to the associated occupational health problems. Although there are still no mechanistic theories universally accepted regarding fibrous minerals and disease, there is evidence which suggests that the flexibility of the mineral with good parting parallel to the fiber length is important. Stanton and Layard (1977) found that long thin fibers without regard to composition were the most carcinogenic in animal studies. They proposed that smaller fibers are readily phagocytosed (assimilated in cells) whereas long fibers are not so readily compromised. Campbell *et al.* (1977) found a distinct difference in fiber lengths and length/width ratios for asbestos and cleavage fragments of amphiboles; and recently Cook (1980) found, in animal implants of the lung, that after an initial clearance of the lung, the fiber concentration stayed constant; cleavage fragments tended to part parallel to and across the major length, whereas asbestos fibers parted parallel to fiber length. It appears therefore that the tensile strength of an individual fiber may be most important in retaining its characteristic length and thus its toxicity.

There has been some debate whether fibers ingested in foods and drinking waters are hazardous. A recent epidemiological study of water supplies in the San Francisco area with naturally occurring elevated levels of chrysotile of 5 to 38 million per liter showed excessive can-

cers above that predicted by socio-economic factors (Kanarek et al., 1980). A similar study has been carried out for municipal water supplies in Quebec by Wigle (1977).

Studies on non-asbestos minerals are less common. Kramer (1977) summarized studies that suggested effects of amphiboles, vermicullite, apatite, gypsum, nemalite, and sepiolite-polygorskite. Webster (1970) noted an effect similar to asbestotic fibrosis in animal studies with nepheline, and Bryson and Bischoff (1967) noted that inhalation of most silicate minerals produces a moderate pulmonary fibrosis.

Attention has been drawn to possible inhalation effects from acid sulphate aerosols which result from acidic precipitation. Hamilton (1979) projected an increased mortality of 3+ deaths per 100,000 population for each microgram of sulphate per cubic meter of air concentration. Mendelsohn and Orcutt (1979) showed that a consistent correlation of excess death could be made to atmospheric sulphate concentrations; they argued further that there appeared to be no threshold level. In Hamilton's data, there does appear to be a level. In either case, the actual nature of the sulphate may be significant. For example, alkaline dusts in carbonate terrains may neutralize acidic aerosols, and the sulphate may react to form a $CaSO_4$ particle. In a different study on the diffusion of SO_2 across Chlorella cells, Spedding et al. (1980) found that only aqueous molecular SO_2 migrated and was toxic. Hence the pH of the medium would be most important in partitioning sulphur species, and the minerals available for buffering reactions would be most important to the pH buffering.

As mentioned previously Burlikov and Michailova (1972) and Mikhailova-docheva (1972) found correlations between soil mineralogy and endemic pleural calcification. They referred to sepiolite and amphibole specifically, whereas Borschchevskii and Konikova (1973) found that apatite and nepheline dusts were correlated with respiratory disease.

It is apparent that outside of occupational health situations, little is known about the health effect of minerals that are distributed in Quaternary sediments. At the same time, very little is known about the nature and distribution of minerals in these sediments. And even less is known about their size distributions and concentrations in adjacent air masses. There is enough suggestive information, however, to be curious enough to study areas where there are known dis-

ease anomalies. Mineralogy, size distribution, and physical properties of individual particles may be key factors in assessing effects. And a sophisticated technology is required to measure these parameters.
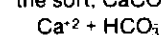
Ingestion

When epidemiological studies cover large geographical areas, hard water is inversely correlated with cardiovascular disease. The correlation does not appear to hold when small groups are considered or when populations are grouped by altitude or by proximity to the seacoast (Safe Drinking Water Committee, 1980). The dividing line between hard and soft water is normally taken as 1.5 meg/L hardness. This is about the concentration for saturation with calcium carbonate if hardness is associated with a Ca^{+2} - HCO_3^- system. There are many other parameters that are associated with hardness, and hardness may merely reflect these parameters.
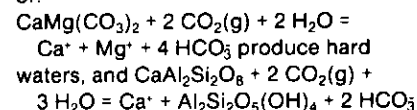
The following categories reflect the possible reasons for correlation of hardness and disease: (1) Harmful ingredients: Soft water can be harmful because of toxic substances present or picked up by the more reactive waters. (2) Indirect protective effect: Hard water can be protective due to the concentration of (trace) substances or the form of the (trace) substance imposed by the hardness factor, (3) Direct protective effect: Hard water may be protective due to the higher concentrations of calcium and/or magnesium, (4) Removal factor: Soft water being more aggressive may leach essential ingredients in a differential manner from foods.

At present, there is no strong evidence to favour one of the categories over another on a universal basis, and it may be that all of the factors are significant.
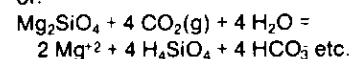
The natural weathering of rocks is responsible for hardness of waters. When calcite or dolomite equilibrate with water, Ca^{+2} and Mg^{+2} is produced, or in soft waters, alumino silicate minerals weather to produce Ca^{+2} and Mg^{+2}. Reactions of the sort, $CaCO_3 + H_2O + CO_2(g) =$

$Ca^{+2} + HCO_3^-$

or:

$CaMg(CO_3)_2 + 2 CO_2(g) + 2 H_2O =$

$Ca^+ + Mg^+ + 4 HCO_3^-$ produce hard waters, and $CaAl_2Si_2O_8 + 2 CO_2(g) +$

$3 H_2O = Ca^+ + Al_2Si_2O_5(OH)_4 + 2 HCO_3^-$

or:

$Mg_2SiO_4 + 4 CO_2(g) + 4 H_2O =$

$2 Mg^{+2} + 4 H_4SiO_4 + 4 HCO_3^-$ etc. produce soft waters.

There are a number of points to consider regarding hardness. (1) Hardness cannot be considered

chemically by itself. There must always be an equal number of negative ions (generally HCO_3^-) for a charge balance. (2) Hardness and bicarbonate are tied to protolyte reactions which normally involve CO_2. Hence directly or indirectly pH becomes the master variable. The previous history of a water (whether subsurface or surface, degree of photosynthetic activity, possible supersaturation and precipitation of oxide and carbonate minerals) may be very significant relative to the concentration and form of other active ingredients. For example, both iron oxides and calcium carbonate are known to be excellent scavengers of inorganic and organic substances. Degassing of CO_2 going from ground to surface water, a raise in pH due to photosynthesis, or increase in temperature would all tend to supersaturate hard waters, but might not supersaturate soft waters. (3) Hard waters will be near equilibrium with $CaCO_3$, and if aerated, will have a pH of about 8,; soft waters will have quite variable pHs but will often be between 6 and 7. This hydrogen ion concentration change of 100 times or more is the largest change of any parameter in the system. (4) Temperature and CO_2 can affect the hardness of a water body to a large degree. CO_2 in surface and groundwaters may range from about 10^{-4} to 10^{-2} atmospheres with a temperature range of about 5 to $30°$ C. Assuming equilibrium with $CaCO_3$, the hardness is given by: $[Ca] = (4 K P_{CO_2})^{1/3}$, where $K = 10^{-4.25}$ ($5°$ C) and $10^{-5.36}$ ($30°$ C). For these ranges, one obtains:

	5° C		30° C	
P_{CO_2} (atm)	10^{-2}	10^{-4}	10^{-2}	10^{-4}
Ca (mM)	6.1	1.3	3.5	0.8

These results show that about one order of magnitude difference in hardness can be obtained from waters saturated with respect to $CaCO_3$. Therefore it appears that knowledge of ground and surface waters may be significant in understanding the evolution of the hardness of a water body.

Schroeder and Kraemer (1974) showed that cardiovascular disease correlated very highly with the Langelier (1936) index. This index, used by water treatment engineers, is the difference between the actual pH and the pH_s of the water saturated with respect to $CaCO_3$, or pH - pH_s. The index is positive for saturated waters and negative for unsaturated waters. The index for waters near saturation with respect to calcium carbonate approximates the logarithm of the degree of saturation with respect to calcium

carbonate. For waters more dilute, other corrections and assumptions have to be made.

There are two very important points regarding epidemiological studies regarding statistical correlation of water parameters and disease. pH and other parameters such as the Langelier index are logarithmic transforms of the concentration data, and this must be considered in statistical evaluation using concentration data; it never has been considered. Secondly, the presence of hard or soft waters is due primarily to the presence or absence of a carbonate mineral. It is questionable whether statistical correlations assuming continuous variables should be used.

The four water factor categories referred to above need comment relative to (1) the ingested quantity from water compared to food, (2) the ability of an ingested substance to adsorb in the body, and (3) competitive interactions. Davis (1980) lists the following points regarding interaction of microelements in mammalian species.

(1) The nature and amount of the element in food resulting from its source and from its processing. Water composition would be important in the cooking of foods, especially vegetables.

(2) The interaction during cell wall adsorption or the competition for sites on body transport ligands. The reactivity due to processing of foods in different waters may be reflected in this step; or a trace element, in excess in water, may compete for a ligand site.

(3) Synergistic or antagonistic interaction within cells and organs of the body.

Schroeder and Kraemer (1974) summarized the relative significance of the amount of elemental ingestion in food compared to water:

Significant water/food uptake (>10%): Mg, Ca, B

Significant water/food uptake (<10%): Si, Ba, Sr, Cr, Cu*, Cd*, Pb* (* - when supplied from plumbing).

Insignificant water/food uptake (~1%): Na, K, Li, Mo, Rb, U, V, Al, Mn

From this summary one might conclude that water is significant for Mg and Ca only. However, this summary only considers a mass input. Many substances were not considered in the analysis, and more importantly, the reactive nature of the substance was not considered; and synergism is not considered.

Food uptake of many trace elements is from vegetables. It is presumed that the metals are strongly complexed as metal-organic and organic substances. Trace metals in hard waters would occur as

inorganic and organic complexes as well as in fine particles; most soft waters would contain a significant amount of free ion, and in acidified waters (pH<6), the trace metals, Al, Cd, Cu, Fe, Pb and Zn would be elevated in concentration over hard waters; but they might pass through the gastrointestinal tract without adsorption. One then can either argue that a trace metal is antagonistic due to its increased concentration in soft waters, or it is labile and is not retained as a necessary element in the body.

The synergistic and antagonistic role, particularly of trace metals, becomes even more complex. Some findings in animal feeding studies are:

(1) Zinc, copper, and iron absorption from the GI tract is a competition for binding sites. Copper deficiency may be due to a lack of the metal or an excess of zinc, a competitive element. Transferrin, metallothionein, and picolinate are probable carrier ligands (Davis, 1980; Sandstead, 1980). Copper deficiency may develop by these mechanisms, and the association may be associated with cardiovascular disease (Klevay, 1980b).

(2) Selenium as selenite ion inhibits mercury toxicity (Parizek et al., 1980) and cadmium toxicity (Whanger et al., 1980). Selenite would be more predominant that selenate in low pH waters for a constant oxidation-reduction potential.

(3) In feeding studies of rats and chickens, Nielsen et al., (1980) showed that Ni has a competitive effect with Fe^{+2} given in food, but has a promotional effect for Fe^{+3}. Ni appears to have a

competitive effect against Cu, and As is required for Zn metabolism.

Picolinate complexes may be very important in intestinal adsorption (Sandstead, 1980). Examination of stability constants along with probable ranges in metal proportions in water give a clue to important metals. These would be Mn^{+2}, Cd, Pb and Zn.

There have been other correlations of water quality parameters with disease. Table I summarizes some examples. Some of these relationships such as the Langelier index and trace metal concentrations can be rationalized to hardness. Others cannot. There also appear to be geochemical factors in the statistics. For example, Crawford et al. (1968) found an inverse correlation with Ca whereas Anderson et al. (1975) found an inverse correlation with Mg. This can be accounted for by the excess of marls in Great Britain compared to Silurian dolomites in Ontario. Similarly Schroeder and co-workers found strong disease correlations with trace metal concentrations and with the Langelier index. His results may reflect the study of softer waters compared to the above studies which considered much harder waters.

There are two examples in Table I which are somewhat different from the other studies. Iwata (1977) showed a correlation of amyotrophic lateral sclerosis on Guam with manganese; he compared drinking waters from wells of soft water, but high in Mn derived from volcanic rocks to hard waters sited in atoll carbonate rocks. There may be an

Table I *Some correlations of disease or effect to properties of water.*

Effect	Water Property	Reference
Excess porphyrins	Lead	Calabrese (1978)
Blood pressure	Sodium	Calabrese and Tuthill (1977a, 1977b, 1979)
Hypertension	Blood lead	Beevers et al. (1976)
Blood lead levels	Lead	Greathouse et al. (1976)
Blood pressure	Lead	Beevers et al. (1980)
Blood pressure	Elevated Na, Cd, low K	Bierenbaum (1976)
Acute hemolysis	Copper	Calabrese and Moore (1979)
Na-induced hypertension	Lead	Calabrese (1979)
Cardiovascular disease	Copper, zinc	Klevay (1980a)
Cardiovascular disease	Trace metals	Craun and McCabe (1975)
Amyotrophic lateral sclerosis	Manganese	Iwata (1977)
Cardiovascular, cerebro-vascular, hypertensive heart disease	Selenium deficiency overall	Shamberger et al. (1975)
Cardiovascular disease	Cadmium, zinc	Masironi (1974, 1976)
Cardiovascular disease	Lithium, vanadium	Voors (1970)
Cardiovascular disease	Ca, Mg, Na, Si, Sr	Sauer et al. (1971)
Arteriosclerosis	$CaCO_3$ saturation, Cd, Sb	Schroeder and Kraemer (1974)
Blood pressure	Calcium	Masironi et al. (1976)
Hypertension, arteriosclerosis	Hardness	Schroeder (1966)
Ischemic heart disease	Calcium	Crawford et al. (1968)
Cardiovascular mortality	Calcium, sodium	Robertson (1968)
Ischemic heart disease Mg in heart muscle	Magnesium	Karppanen and Neuvone (1973), Marier et al. (1979), Hewitt and Neri (1980)

analogy for more ancient volcanic rocks. Shamberger et al., (1975) found an inverse correlation between disease and selenium content; this correlation can be related to hardness through pH as the important selenite ion is more predominant at lower pH.

Acidic precipitation may or may not have an effect on the hardness - cardiovascular disease statistic. Acidic precipitation may produce waters higher in trace metal concentrations, but there may well be an increase of Ca and Mg due to the increased rates of weathering. Fuhs (1979) found waters of 7 mg/L Cu and 0.2 mg/L Pb in metal plumbing systems consuming soft waters in the Adirondack region of New York; springs contained similar concentration levels presumably due to trace metal exchange

with soils. In England and Scotland, corrosion of lead from old plumbing fixtures in soft water regions has been correlated with high blood lead level, elevated blood pressure, and hypertension (Beevers et al., 1980; Shaper et al., 1980).

In summary, there appears to be a strong correlation between low cardiovascular disease and related symptoms and hard water concentration. The actual causative factors are not clear and may be due to nutritional requirements of Ca and/or Mg, excess or deficit of trace metals, a trace metal inbalance developing antagonistic effects, or some other factor. Hardness of waters results from the history (predominantly ground water compared to surface water) and the contact or lack of contact with carbonate minerals.

Hard Waters and Quaternary Sediments of Eastern Canada

The last glaciation involved two major glacial sources. One involved carbonate rocks of the Hudson Bay region and covered most of Ontario and some of Western Quebec, and the other glacial advance involved non-calcareous rocks from Quebec-Labrador and covered the Maritime Provinces and Eastern Quebec. Therefore Quaternary sediments on alumino-silicate rocks should exhibit hard waters for areas of Ontario and soft waters for areas of the Maritimes and Quebec. Figure 1 is a map with the surface boundary between the two kinds of sediments shown, and superimposed upon this map is the calcite saturation index (CSI) for major stream crossings. The CSI is defined as the negative logarithm of the degree of saturation with
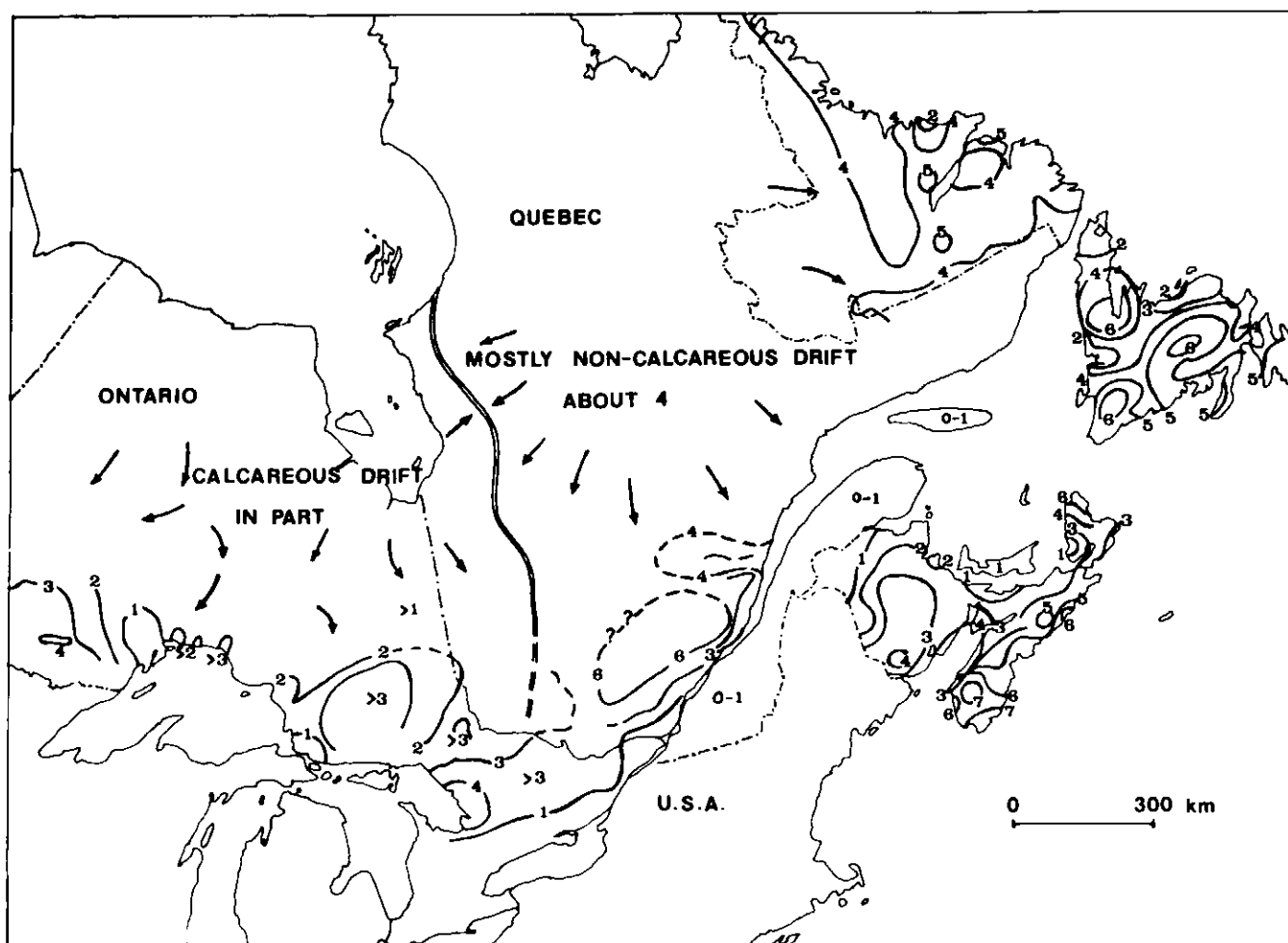

Figure 1 *Calcite saturation index (CSI) for Eastern Canada. Hard waters are shown by values of zero or less; soft waters are shown by numbers of 3 or greater. Ontario data compiled from McMaster University stream surveys; Maritimes data from Clair et al. (1980); Quebec data from Environment Canada (1980) and*

McMaster University. The double line separates surface exposures of glacially derived sediment with some carbonate mineral content (Ontario) from material with essentially no carbonate content (Quebec). Arrows show direction of glacial movement. Published with permission of the National Research Council of Canada.

Diagram after Fig. 3-15 in Acidification in the Canadian Aquatic Environment: scientific criteria for assessing the effects of acidic deposition on aquatic ecosystems (NRCC report 18475, 1981).

respect to calcite. Therefore the higher the value of CSI, the softer the water. A value of zero would represent saturation and negative values would represent supersaturation with respect to calcite. One can see a fairly close correlation of the CSI values with the two different Quaternary sediment masses. There are only a few isolated areas in Ontario with high CSIs (soft water), whereas large portions of Eastern Canada have very soft waters. Exceptions are near seacoasts where calcareous marine sediments cover coastal lowlands (St. Lawrence, etc.) and where Paleozoic sediments are found. There would be other variations due to local lithology, ground water effects etc., if viewed in more detail.

In Ontario, low CSI values (hard waters) are typically found in lacustrine deposits and some outwash deposits, whereas high values (soft waters) are found often in ground moraine.

This gross correlation of water quality to glacial history of the region suggests other geochemical and Quaterary sediment-bedrock studies. For example, it would be most interesting to know the correlation of manganese water concentrations with volcanic rock types in low hardness areas and the association with disease. It might also be of interest to know the chrysotile concentration of these waters if they percolate through sediment derived from serpentinized volcanics (Schrier and Taylor, 1980). Or if magnesium is an important nutritional factor, it would be of value to know the distribution of Mg in unconsolidated sediments.

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