Report on Bioaccumulation of Elements to Accompany the Inventory of Radionuclides in the Great Lakes Basin

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BACKGROUND

In December 1997, the International Joint Commission (hereafter referred to as The Commission or (IJC) released the report of its Nuclear Task Force (also referred to as the Task Force), *Inventory of Radionuclides for the Great Lakes* (hereafter referred to as the *Inventory*). This report presented data on the sources of various radioactive isotopes and used a material balance approach to organize this information to permit various types of assessments with respect to the movement and distribution of radionuclides within the Great Lakes. One component of such a material balance is the radionuclide burden within biota of the various biological communities within the Great Lakes. Estimation of the biological burden of various radioactive elements requires a consideration of the exposure to and uptake, metabolism, excretion, and transfer of these radioactive elements within and among organisms. To address these issues, the Task Force undertook a study of bioaccumulation of radioactive materials from environmental media and substrates and biomagnification of radioactive materials within biota, food chains, and biotic assemblages. The studies undertaken examined a large number of data sets that might be suitable for determination of bioaccumulation and biomagnification parameters for various radioactive forms of elements. At the time of preparation of the *Inventory*, several of the reviewers indicated that analyses for bioaccumulation and biomagnification should be separately issued by the Commission as reference documents. This report, referred to as the *Bioaccumulation Report*, is a response to that suggestion.

INTRODUCTION

An important component of the environmental transport and distribution of elements is their cycling through biological compartments of ecosystems. This is also one of the most difficult processes to study. Even for the most heavily studied elements in biological compartments (*i.e.*, carbon, nitrogen, or phosphorus), the research has been painstaking, taxed the ingenuity of investigators, pushed to the limits the state-of-art of instrumental and chemical methods, and raised more research questions than it answered. Because the cycling of elements integrates organismal chemistry and biology, this subject plays a role in predicting the effects of elements on organisms, biological communities, and ecosystems.

The cycling of elements through biological compartments does not "theoretically" depend on whether a nuclide is stable or radioactive. A radioactive isotope cycles the same way as the stable isotope of the element. Thus, many studies on how elements cycle use radioactive isotopes as tracers. The quotation marks around *theoretically* suggest that perhaps the cycling of elements does sometimes depend on some particular property of an isotope, although it is unclear that nuclear stability is the property of interest. Differences in biological properties occur among isotopes of elements of low atomic weight. The substitution of deuterium (^2H) for hydrogen in some molecules changes the physicochemical and biological properties of the molecule. Deuterium often increases the toxicity of a compound. Likewise tritium (^3H) also when substituted for hydrogen in molecules changes physical, chemical, and biological properties of the molecule.

The radioactive decay of a nuclide can produce a new nuclide of a different element. When that occurs, the new nuclide begins to cycle according to the chemical identity of the new element.

PART I

ANALYTICAL CONCEPTS

Some background concepts of an analytical nature will assist in understanding the materials presented throughout the report.

LATITUDE ADJUSTMENT TECHNIQUES FOR NUCLIDES IN FALLOUT

In the early work on deposition of radioactive particles from the atmosphere to the surface of the earth, a simplified model was developed and used by atmospheric scientists and exploited by UNSCEAR. This model treats earth as a sphere (actually it is more of a prolated ellipsoid because of the configuration at the North and South poles) of radius 12,800 km. The surface of the sphere is divided into latitude bands of 10° latitude width from North Pole to South Pole. The designation of latitude bands is north or south of the Equator, which by convention is designated 0° latitude. The Equator represents the latitude of maximum spherical circumference. The numerical designations of either a north latitude or south latitude range from 0° to 90°.

Nuclear scientists have determined that deposition of atmospheric radioactive material, because of factors ranging from the locations of nuclear sources to the axial tilt of the earth relative to the sun, that 10° latitude bands are the smallest statistically useful scale to resolve the deposition data. The Great Lakes occupy parts of the latitude bands from 40°N to 60°N. Their surface area for deposition is 24.6×10^4 km^2, the world-wide area in that latitude band for deposition based on the spherical assumption is 67.2×10^6 km^2. The UNSCEAR model assumes uniform deposition over the sphere; therefore, the fraction of the global deposition falling on the Great Lakes surface available area is 0.00366. The drainage basin is 49.6×10^4 km^2, but it extends below 40°N and above 60°N. Because the increased area for deposition is small compared with the area of global deposition, one need not adjust for the extended area outside of the two boundary latitude bands. Thus one can use a simple proportionality to obtain the increased deposition associated with the drainage basin relative to the Lakes. The fraction of the global deposition falling on the Great Lakes drainage basin is approximately 0.0074. These deposition data are then corrected for radioactive decay of the isotopes.

STATISTICAL APPROACHES — COMPOSITIONAL DATA

Statistical data for substrates or media that contain, receive, or export multiple isotopes require multivariate statistical techniques. The data do not represent the entire chemical composition of the substrate or media but, rather, are some unknown fraction of the same. This raises the issue of the applicability of what Aitchison (1986) called *compositional data*. While one assumes that the sum of all the elemental parts is 100% and the sum of fractions is unity, in the actual analytical situation the variability of one or more components may be artificially restrained by this model. Most data sets on isotopes or chemicals, especially those used to consider biological cycling of elements through biota and biotic communities are compositional data sets. They usually have, however, enough unspecified or unmeasured subsystems to permit a relaxation of any statistical restrictions associated with compositional data, especially when assessing correlation or analyzing variance.

Although many radionuclides occur in the discharges from various nuclear sources to the Great Lakes, not every element has radionuclides that will bioaccumulate and affect biota of the Great Lakes. The Task Force, in its work on the development of inventories of "all" radionuclides found in the Great Lakes, recognized the need to bound its inquiry. It addressed only those elements whose radionuclides require inventories for biological compartments. Such elements usually share one or more of the properties listed in Text Box 1. In reviewing the material in Text Box 1, the Task Force notes a special concern with respect to the half-life of a radionuclide: beyond the separate importance for studying ^{131}I (half-life 8 days), the Task Force has chosen to study radionuclides with half-lives greater than 14 days. The Task Force wishes to acknowledge some problems, encountered in the preparation of this report, which could not be resolved based on currently available data.

The Task Force addressed in a limited way the following elements: arsenic, cadmium, copper, and mercury. Data on the bioaccumulation of these elements exists from which to develop bioaccumulation factors, but radioactive isotopes of these elements are not major concerns in the Great Lakes. Given the known toxicity of these elements as well as the toxicities of osmium, tellurium, and thallium, one might consider it strange that the bioaccumulation tables in this report do not include many entries. The Task Force had no elemental data for osmium, thallium, and tellurium. Other unusual elements are tantalum and antimony. *Tantalum has not been documented as present in aquatic biota.* Antimony is toxic to aquatic organisms, and its radionuclides are known fission products, which have been detected in the Great Lakes, but *antimony's presence is freshwater aquatic organisms has only very rarely been documented.* Table 1 lists elements that were considered in this work.

TEXT BOX 1
PROPERTIES OF RADIONUCLIDES THAT WOULD DIMINISH THE NEED TO ESTIMATE BIOACCUMULATION AND BIOMAGNIFICATION PARAMETERS

(1) **SHORT-LIVED RADIONUCLIDE WITH HALF-LIFE OF LESS THAN ONE (1) WEEK.** The monitoring of most radioactive releases tracks radionuclides with half-lives of at least eight (8) days, mainly because of this is the half-life of ^{131}I, a radionuclide of great importance in health considerations of humans. Those radionuclides with half-lives less than eight (8) days do not retain their original nuclear identity long enough to cycle through biological compartments, even as radioactive versions.

(2) **GENERATION OR PRODUCTION OF THE RADIONUCLIDE OCCURS UNDER CONDITIONS THAT ARE UNRELATED TO NUCLEAR INPUTS TO THE GREAT LAKES.** For example, some radionuclides only arise in nuclear reactor experiments or laboratory situations without environmental discharge, while other nuclides occur only in "sealed sources" and are not released to the Great Lakes.

(3) **THE NUCLIDE IS PRODUCED COSMOGENICALLY, AND ITS BIOLOGICAL INVENTORIES WERE PREVIOUSLY ADDRESSED IN THE *INVENTORY* REPORT.**

(4) **MOST OF THE NUCLIDES OF AN ELEMENT ARE STABLE AND THE END PRODUCTS OF A SELECTIVE DECAY CHAIN.** Here inventories would examine nonradioactive situations mainly rather than radioactive situations. Geochemists and others may have use of these stable nuclides for reference purposes or to assist in developing the inventories for the radionuclides of other elements. In those cases, the element is discussed, but no inventory is presented.

(5) **THERE IS INADEQUATE EVIDENCE OF THE NUCLIDE'S BIOLOGICAL UPTAKE.**

(6) **THE LIMITED INFORMATION ON THE BIOLOGICAL CYCLING OF AN ELEMENT DOES NOT SUPPORT ESTIMATES OF ITS BIOACCUMULATION FACTORS.**

TABLE 1
CLASSIFICATION OF ELEMENTS FOR REQUIRING ESTIMATES OF BIOACCUMULATION AND BIOMAGNIFICATION PARAMETERS FOR BIOTA OF THE GREAT LAKES

I Elements whose radionuclides do NOT require estimates of bioaccumulation and biomagnification parameters for biota of the Great Lakes:

Arsenic	Bismuth	Cadmium	Copper	Fluorine	Gallium
Germanium	Gold	Hafnium	Indium	Iridium	Mercury
Osmium	Oxygen	Palladium	Platinum	Rhenium	Rhodium
Scandium	Tantalum	Tellurium	Thallium	Tungsten	

II Elements whose radionuclides do NOT require estimates of bioaccumulation and biomagnification parameters for biota of the Great Lakes but for which analyses may be needed in developing and discussing the bioaccumulation and biomagnification processes of other elements in Great Lakes biota:

Aluminum	Boron	Bromine	Calcium	Magnesium	Silicon
Titanium	Selenium	Sulfur	Vanadium		

III Elements whose radionuclides have an assumed presence in biological compartments of ZERO:

Antimony	Argon	Neon	Krypton	Xenon

BIOACCUMULATION AND BIOMAGNIFICATION

The terms *bioaccumulation and biomagnification* refer to the uptake of chemicals by organisms and their retention in amounts that may exceed the levels present in the source materials. *Bioaccumulation* is a more general term, with *biomagnification* describing a special type of bioaccumulation usually associated with dynamics of organismal food webs and the cycling of chemicals through biotic assemblages and ecosystems. Furthermore, biomagnification has been used more often in discussions of organic chemicals than radionuclides.

Bioaccumulation can be assessed directly or indirectly. Direct measurements quantify the residue or elemental/chemical content of some substance in tissue, for example, analytical determination of the content of ^{40}K in a tissue. Indirect measurements examine some indicator of bioaccumulation, measure the indicator, and use statistical correlations or calibration equations to relate the measure to the accumulated material, for example, measurement of chlorophyll *a* in algae as a surrogate for productivity or biomass production.

It is sometimes possible to use mathematical models and simulations to predict bioaccumulation and biomagnification. The simulation models emphasize the cycling of chemicals through biotic assemblages (communities, trophic levels) and ecosystems in somewhat generalized form, usually as simplified foodwebs or energy flow systems.

Simulation techniques usually work better with organic chemicals than with radionuclides despite the fact that some simulations were originally developed for radionuclides. The success of the simulation techniques with chemicals mainly reflects their dependency of many bioaccumulation processes on the size of an organism's lipid reserves with the bioaccumulated chemicals depositing selectively in lipid tissues. Several physicochemical (*i.e.*, quasi-thermodynamic) properties of chemicals express this partitioning of a chemical between lipid and aqueous tissue reserves rather well, the best known one being the "octanol–water partition coefficient." Radionuclides that show an affinity toward lipids include those whose stable nuclides are parts of lipid compounds (*e.g.*, carbon) and those that form lipid-soluble compounds, notably metals which form organometallic compounds through such mechanisms as methylation (*e.g.*, mercury, tin, arsenic, selenium).

Simulation techniques also work well with the known metabolic pathways and mechanisms for specific compounds. Compounds of interest include the vitamins, hormones, and small molecules that act as *cofactors* in metabolism. These compounds either contain an unusual element (*e.g.*, vitamin B_{12} has cobalt, thyroxine has iodine) or require the presence of a special element for activation (*e.g.*, magnesium, iron, manganese, molybdenum), and the unusual or special element can easily occur as a radionuclide.

Bioaccumulation processes are dynamic. They may occur over time periods that appear to be essentially "infinite" relative to the expected life-span of an organism. However, the limitations of organismal size specify the number of tissue sites or of tissue capacity to carry chemicals coupled with physiological processes of detoxification and internal processes of tissue repair. That assures that bioaccumulation would reach a steady state. This "steady state" may be an academic concept, since very few bioaccumulation processes have received sufficient study to show that the steady state has been reached. Many factors can confound the observation of a steady state and are presented in Text Box 2 and discussed individually.

TEXT BOX 2
FACTORS AFFECTING OBSERVATION OF STEADY STATES
IN BIOACCUMULATION PROCESSES

(1) **ORGANISMAL GROWTH, REPRODUCTION, AND DEATH**

(2) **STORAGE IN SPECIAL MATRICES**

(3) **EXPOSURE HISTORY OF BIOTA TO VARIOUS ISOTOPES OF A GIVEN ELEMENT**

(4) **GEOCHEMICAL AND METABOLIC "COHERENCE" (DEPENDENCE OF THE BIOLOGICAL AND CHEMICAL BEHAVIOR OF AN ELEMENT ON THE PRESENCE AND AMOUNT OF OTHER ELEMENTS**

Organismal growth increases capacity to bioaccumulate a given nuclide. Depending on past exposure, organismal growth may also include adaptation to an element not previously experienced. DeFilippis and Pallaghy (1994) refer to "a considerable induction of tolerance" in algae chronically exposed to heavy metals and the observations of reduced levels of toxicity under certain conditions of metabolic activity, notably in selective carbon and nitrogen assimilation processes. Organism reproduction also provides a mechanism for a parent to pass some of its accumulated material through food to an offspring, lower its own body burden, and free up existing or create more capacity to accumulate more material.

Is a tissue residue for a substance, even if a lethal level, the "steady-state" level or was passive uptake the method of bioaccumulation despite organismal death? Biological variability in susceptibility among organisms both intraspecifically and interspecifically creates a range of observed or measured "fatal" levels, which may not clearly indicate that some maximum observed "lethal" level is *the* steady-state level. Emerson and Hesslein (1973) added ^{226}Ra to Lake 227 of the Experimental Lakes Area (ELA) at Kenora, Ontario, and followed the distribution of both the radium and radon gas daughter nuclide in various components of the lake. They reported that the uptake of radium did not differ between live and dead algae, suggesting at least for this radionuclide, the importance of passive adsorption mechanisms in the biouptake processes. Further, their experiment suggests the importance of the particulate matter from dead organisms as a binding material for radionuclides and, thus, the potential to observe continued biouptake long after an organism has died.

By incorporating accumulated material into a special matrix that either renders the material inert in storage or hastens detoxification, the organism may add capacity to accumulate. Pentreath et al. (1980) studied the accumulation of ^{237}Pu by seaweeds such as *Fucus vesiculosus* and *Fucus serratus*. They showed that accumulation occurred mainly by surface adsorption with the bulk of the plutonium found in a very thin outer surface layer of the laminae. Early studies by Harvey and Patrick (1967) on the uptake of ^{137}Cs and ^{65}Zn by diatoms showed that the surface to volume ratios of the cells correlated with the bioaccumulation factors to a greater degree than the taxonomic characteristics of the diatoms studied. The matrix of mucoproteins and glycosidic residues on the organism surfaces provide the storage mechanism. Since these materials are water soluble, the investigators noted that the radionuclides washed off the organisms rather easily.

The background concentration of the substance in the source materials may dictate the extent of accumulation. Titanium usually has a very low uptake in biological organisms. Yan *et al.* (1989) found very low residues of titanium in the zooplankton species of Bat Lake and very high residues of titanium in the zooplankton of Horn-2 Lake. These lakes, in the Canadian Shield region of northwestern Ontario, had respectively, very low and very high titanium content of source material of the lakes' basins: the trap rock and water. In fact, Bat Lake had such a low mineral content in the source materials that most of the elements accumulated in the zooplankton species reflected the rather impoverished environment. By contrast, the zooplankton in Horn-2 Lake, a rather alkaline lake, had considerably elevated levels of calcium, copper, aluminum, magnesium, strontium, barium, zinc, and cadmium, reflecting the nature of the source materials.

The accumulation of an element very often depends on another element or suite of other elements, as well as its own levels in various environmental compartments. Hutchinson (1975), in his *Treatise on Limnology, Volume III,* noted that one of the more interesting problems in understanding the chemical ecology of lake macrophytes is the "unusual" array of accumulated elements that are sometimes observed. Does the accumulation of and value of the steady-state residue for some unusual element, such as titanium, depend on the presence of calcium, magnesium, aluminum, copper, etc. It is known that uptake of barium depends on calcium, but the unknown nature of the mechanisms for such "assisted" or "dependent" uptake complicates the problem.

Bioaccumulation factors

The fundamental parameter expressing bioaccumulation is the *bioaccumulation factor,* a dimensionless ratio of the concentration, activity level, or similar entity of a chemical within a given tissue to its comparable concentration, activity level, etc., in the source material to which the tissue is exposed. The bioaccumulation factor can be extended to describe relative distribution between two biological materials, a biotic source material and a second biological substrate exposed to the source material. If this ratio exceeds unity, one has "biomagnification" and the bioaccumulation factor becomes a "biomagnification factor." The usual situation compares the ratio of bioaccumulation factors for a nuclide for two species, a consumer species and a consumed species.

Reference elements

Geochemists sometimes choose a "reference" element for comparison when studying the geochemical relationships of chemicals and elements to each other. The technique often works well with bioaccumulation, but does not have a significant history of use for biomagnification. In fact, the Task Force does not recommend its use for biomagnification, unless there is evidence of the biomagnification of several nuclides by the *same or very closely related mechanisms, and that the biomagnified nuclides are geochemically "coherent."* "Coherence" is a geochemical term for some entity accompanying something else in a mechanistic manner such that inferences about the thing and that which it accompanies treat both entities identically. Such coherence can often overrule some of the obvious behavioral differences that would be ascribed to the entities based on chemistry, biology, or physics alone, if they were treated separately under the circumstances described.

There are many choices of reference element, but most currently used choices fit one of two categories: another element in the same family of the Periodic Table as the element of interest, or an element that reflects knowledge of the source materials for the nuclides being accumulated. In the first choice, the comparison to the reference element examines the chemistry of the family of the Periodic Table to understand the extent to which a given element follows the behavior of the reference element. In the second choice, the comparison examines the extent to which the element of concern follows the known behavior of the reference element in various types of source materials and substrates.

For the second choice consider the nature of soils. Elements that have limited biological mobility and would likely remain in soil in preference to uptake by an organism might show a certain mutual correlation. Titanium and aluminum tend to have limited biological mobility, although the Task Force has noted data that significantly challenge this "common wisdom" and have served as reference elements when examining the bioaccumulation of elements from mineral, soil, and related materials. When relating radionuclides to each other from source materials, the primordial natural radionuclide of potassium, ^{40}K, sometimes serves as a reference element.

A third choice, pointed out by Dr. Ursula Cowgill (personal communication) examines an element *not known to be present in any of the substrates, media, or source materials* as the reference. Such an element has complete accountability in all materials *because its levels are those that have been quantitatively added to the system by the investigator for reference purposes.* The comparisons are for material-balance calculations with quality-assurance checks inherent in the knowledge that all sources and amounts of the reference element are under the investigator's control and quantitatively known.

The use of a reference element changes the calculations and interpretations of bioaccumulation and biomagnification factors. First, the reference element approach converts the calculation of a bioaccumulation or biomagnification factor from a single geochemical ratio of a given element in two different source materials to a ratio of two geochemical ratios of the given element to reference element in each of the two different source materials. This new calculation procedure changes the interpretation of the bioaccumulation factor from the relative enrichment or rejection of the given element between the two different source materials to relative enrichment or rejection of a coherent pair of elements in the two different source materials. The incorporation of a reference element into the calculation may suggest the presence or absence of a commonality of the environmental distribution of the element being studied with the known environmental distribution of the reference element. The resulting bioaccumulation factor is sometimes referred to as the *enrichment factor*, because it compares behavior of an element with respect to a reference element in the uptake process to show the pattern of "enrichment" (increasing uptake relative to the reference) or "rejection," sometimes referred to as "depauperation" (failure to transfer to a new phase relative to the source materials).

TEXT BOX 3
BIOACCUMULATION FACTOR AND BIOMAGNIFICATION FACTOR FORMULAS

Without reference elements:

[1] Bioaccumulation factor for a nuclide

$$= \frac{\text{Activity or concentration of nuclide in organism}}{\text{Activity or concentration of nuclide in source material}}$$

[2] Biomagnification factor for a nuclide

$$= \frac{\text{Activity or concentration of nuclide in consumer species}}{\text{Activity or concentration of nuclide in consumed species}}$$

With reference elements:

[3] Bioaccumulation factor for a nuclide

$$= \frac{\text{(Activity of nuclide in organism)/(Activity or concentration of reference element in organism)}}{\text{(Activity of nuclide in source material)/(Activity or concentration of reference element in source material)}}$$

[4] Biomagnification factor for a nuclide

$$= \frac{\text{(Activity of nuclide in consumer species)/(Activity or concentration of reference element in consumer species)}}{\text{(Activity of nuclide in consumed species)/(Activity or concentration of reference element in consumed species)}}$$

The formulas for the various types of bioaccumulation factors are given in Text Box 3. The formulas do not depend on the stability of the nuclide. Obviously "activity" data will apply to radioactive materials, but the formulas are generic, since specific activity is defined as proportional to concentration.

Reference elements and Great Lakes data

When the source material of a nuclide is water, minerals, soils, or sediments, the reference element approach sometimes gives a greater bioaccumulation factor by suggesting possible geochemical relationships between elements to improve the understanding of an organism's uptake, rejection, or retention of a given element. Some marine geochemists used this approach to separate bioaccumulation rates and mechanisms for elements found in surface layers of the oceans from those in deeper layers of the oceans. The studies the Great Lakes usually do not have sufficiently rich data collection protocols to exploit the more advanced ideas in the reference element approach, but the Task Force has attempted this approach in a few instances.

Quality assurance provisions for biouptake data

Because of the variability in quality and form of the data for bioaccumulation and biomagnification, and because the bioaccumulation and biomagnification factors can be used for estimating the nuclide inventory of a biological compartment, the Task Force has listed a set of criteria for rating the quality of the data used. Text Box 4 summarizes this information. The most reliable data would fit a profile of all "Yes" answers to the questions, although the listing is not exclusive. Those data that a fit a profile of only a few "Yes" responses would engender much less confidence as to quality and usefulness.

A special note about the fifth question in Text Box 4. The Task Force does not expect that the information provided has associated with it a legal "chain of custody," as sometimes prescribed in protocols for research and monitoring that must meet legal or regulatory requirements. Rather the Task Force wishes to know enough about the collection, processing, handling, and analysis of materials and the subsequent reporting of results to assure confidence that the data can meet some tests of quality in later analyses of bioaccumulation and biomagnification processes and the development and assessment of radionuclide inventories.

The sixth question in the Text Box 4 on chemical and biological matrices refers to a practical problem of available reference samples of biological materials for quality assurance purposes. Some reference material is supplied by the National Institute of Technology (formerly: United States Bureau of Standards). The matrix problem in general requires careful review before using elemental data obtained from many substrates. Other reference samples have been developed cooperatively with the United States Environmental Protection Agency on radionuclides in a variety of substrates, some of which are of biological origin.

TEXT BOX 4
QUESTIONS TO ASSESS THE QUALITY OF DATA USED TO EVALUATE BIOACCUMULATION AND BIOMAGNIFICATION

(1) Is there a complete identification of the biological sample being examined? If the sample is tissue from an organism, is the type of tissue or part clearly and completely identified, including the organism source and its taxonomic name?

(2) If the biological description is for a "mixed" material (*e.g.*, periphyton or plankton), is the description sufficiently ecologically clear to permit interpretation of the observed bioaccumulation and biomagnification parameters?

(3) Are the data available on a "dry weight" basis. If data are on a "wet weight" basis, are the per cent moisture and the per cent "ignition loss" given to permit the calculation of results on a dry weight basis?

(4) For an element, is (are) the valence state(s) given? If not, are the analyses reported on total element basis?

(5) Are the conditions of handling of the specimens well documented, including: irradiation times or exposure time, sampling protocols if the material were taken from the field, methods to prevent cross contamination of specimens from the materials of handling, preservation methods, analysis methods (counting, gamma spectra, etc.)?

(6) Are the source materials and their matrices from which samples were collected clearly identified (*e.g.*, water, trap rock, sediment, air–water interface, foams and sprays, sediment–water interface)?

Biotic needs for bioaccumulation data

There are several data needs related to bioaccumulation. First is a need for biological productivity data. Productivity data converts concentrations and activities of nuclides into mass balance parameters and allows one to address whole-organism data as well as ecosystem and community data by considering the absolute rather relative cycling of selected isotopes.

For lakes, one needs data for plankton species (phytoplankton and zooplankton), benthos, and pelagic fishes. For rivers and tributaries that enter, connect, or leave lakes, one needs the comparable data for benthic algae, macrophytes, and macroinvertebrates. Finally, there are terrestrial species that provide a link between elements that cycle through aquatic biota and terrestrial biota. These include fish- and invertebrate-consuming birds, reptiles, and mammals (herein considered as "wildlife"), which spend part of their time in lake and river habitats but are not totally aquatic species.

The major data deficiencies occur for lake macroinvertebrates, both benthic and planktonic, river plants and animals, and the terrestrial wildlife species. Since 1985, several government research laboratories, mainly in Canada, have sought to close these data gaps. In many instances the focus of the research was on Arctic species or biological assemblages in national parks or remote areas, not specifically the Great Lakes. Nevertheless, the climate zones, latitude bands, species distributions, and often a fortuitous coverage of radionuclides from residual atmospheric fallout and weapons testing have enabled of the Task Force to address Great Lakes biotic compartments. Of particular value are data for species of *Daphnia*, which are important in the Great Lakes.

Limitations of elemental composition data

The simplest measures of bioaccumulation derive from the elemental composition of a tissue or organism and compared with the elemental composition of its food, water, or host environment. However, the comparisons of data on elemental composition of several biological samples or substrates do not always quantify the bioaccumulation processes.

One consideration is that very few residue measurements are taken as functions of time, and therefore, the *state of bioaccumulation does not infer the rate of bioaccumulation*. Further, the bioaccumulation data may reflect a history of organismal exposure, which is unknown to the investigator. This occurs routinely in migratory species or organisms that experience rapidly changing environmental conditions.

Organisms also can try to control much of their physiology and behavior to maintain *homeostasis* (an operational range of states that permit the organism to function successfully and minimize stress). This requires that an organism exert some control over its elemental composition despite fluctuating conditions of elemental scarcity or excess and changes in external physical environment (*i.e.*, temperature, pH, alkalinity, salinity, flow, and dilution). In maintaining homeostasis, organisms may modify considerably their own exposure history to the elements which they accumulate.

Assimilation efficiency

Calow and Fletcher (1972) pioneered a technique called the "assimilation efficiency," which relates the uptake of a chemical to the assimilation of carbon (a measure of new protoplasm production). The method designates the carbon assimilated from food sources as the reference element to which all other accumulated elements are compared. Note that the method only considers that part of the geochemical pool of carbon that goes into protoplasm. Other reference element methods do not make this distinction but, rather, make their comparisons with respect to the total pool of the element in a given medium, not a specific fraction of that element in a given medium. The choice of the element's entire geochemical pool is sometimes unavoidable because the studies cited had no methods for measurements of the various fractions of the element's pool that would allow a more refined choice. Many *transfer factors*, which relate elements found in certain organisms to the amount retained in consumer species, *assume a 50% utilization of available carbon*. This assumption occurs in models using marine species, since that data base is large.

^{14}C is a tracer of assimilated carbon. The method assumes a constant ratio of radioactive carbon to total carbon (complete mixing of the isotope pool), thus assuring the ratio of radioactive carbon to assimilated carbon is a constant. For a second element, a radionuclide version is used under other circumstances, which assures that element is also a constant fraction of its geochemical pool. The ratio of the activities of the two reference radionuclides equals the ratio of the elemental masses in the appropriate geochemical pool of the substrate being studied. The second isotope is typically ^{51}Cr, which is assumed to have no assimilation or protoplasm purpose. The retention of the radionuclide of chromium thus presumably measures some passive biouptake and storage mechanism. The geochemical ratios of $^{14}C/^{51}Cr$ in organismal food and fecal materials are determined and the assimilation efficiency obtained by difference.

Nicholas Fisher and his coworkers (Fisher et al. 1983; Luoma et al. 1992; Wang and Fisher 1996) have explored this method extensively with ^{51}Cr, ^{57}Co, ^{65}Zn, ^{75}Se, and ^{110}Ag with marine molluscs (*e.g.*, *Mya arenaria*, *Mytilus edulis*) and various species of marine algae. Fisher could derive regression equations between ^{14}C and some radionuclides accumulated in selected species, but in a few cases, especially ^{110}Ag, such regression equations could not be derived. Regression equations enhance an understanding of the biouptake processes in that one can see which elements *correlate* with carbon assimilation, but the absence of such equations offers *equally important information on elements that do not correlate with carbon*. For example, the environmental movement of silver is not well understood; thus, it is useful to know that the cycling of this element does not show an association with the assimilation and use of carbon in the nutrient pool.

One advantage of the assimilation-efficiency method is its consideration of growth and production of new protoplasm into the analysis, while treating dynamically the uptake, excretion, and retention of a radionuclide. The mathematical equations describing the method can treat several kinds of experimental data, including data from static and dynamic experiments and absolute concentration data as well as kinetic data on rates of change of concentration with time. The mathematical equations appear in Text Box 5.

TEXT BOX 5
RELATIONSHIP BETWEEN ASSIMILATION EFFICIENCY AND BIOACCUMULATION FACTORS

[1] $$\text{Assimilation Efficiency } (AE)_C \text{ (for carbon) (\%)} = \left| 1 - \frac{(^{14}C/^{51}Cr)_{feces}}{(^{14}C/^{51}Cr)_{food}} \right| \times 100$$

[2] $$\text{Assimilation Efficiency } (AE)_X \text{ (for element } X\text{) (\%)} = \left| 1 - \frac{(X/^{14}C)_{feces}}{(X/^{14}C)_{food}} \right| \times 100$$

[3] $$\text{Bioaccumulation factor (relative to food)} = (X/^{14}C)_{tissue}/(X/^{14}C)_{food}$$

[4] $$\text{The accumulation of element } X \text{ in tissue} = X_{stored} + X_{assimilated}$$

The assimilation-efficiency method addresses an aspect of the transport process of fecal pellets of organisms excreted into the water column and moving to lake sediments. The settling of particles in the water column, of which the fecal pellets of organisms are an important component of the particulate matter, is a major transport mechanism of many elements to lake and ocean sediments. The marine studies far exceed in number and scope the studies of the freshwater systems. Most of the studies of vertical transport of particulate matter in lakes examines diurnal variations in plankton movement. The estimates of element fluxes to sediments are far more advanced for marine programs than for lake programs, but the method can contribute to an understanding of the settling of particulate matter in lakes.

The choice of chromium as a reference element in the excretion process, because chromium ostensibly has no role in the production of protoplasm, is questionable. Chromium has a role in vertebrate metabolism (see *Chromium (1975)*, a report of a committee of the United States National Academy of Sciences), but its role in invertebrate and plant metabolism is unknown. In the literature on assimilation efficiency, statistical arguments suggest that one can use chromium data to cross check the incorporation of carbon from nutrient materials from a chromium to carbon ratio.

A lack of comprehensive data sets

Hutchinson (1975) noted the wealth of data on the elemental composition of various species, but the completeness of the data was another issue. Most of the available radionuclide data refer to agricultural crops and products because of the requirement to monitor the food supply for direct contamination by radionuclides and concerns about the transfer of nuclides from the atmospheric fallout associated with atmospheric testing of nuclear weapons during the 1950s and 1960s.

Two kinds of data typify the radionuclide studies of foodstuffs: (1) radioactivity measurements of crops in farms near the site of a nuclide discharge to the environment, especially grass used in animal feeds, the milk content of local dairy cattle, and sometimes even the farm animals themselves; and (2) radioactivity measurements on crops that have been deliberately grown either on radioactively contaminated or amended soil or treated with radioactively contaminated or amended fertilizers. Periodically, selected government agencies screen the produce and meats at selected stores at locations either near areas receiving nuclide discharges or that receive their produce from areas near places of nuclide discharges.

A third kind of data comes from "geobotanical prospecting." Certain plant species are called "accumulators" because they selectively accumulate certain elements and enables a person to infer a geological resource by virtue of the *presence* of the plants. Geobotanical prospecting was used successfully to locate exploitable sources of uranium, lithium, gold, and silver.

Prior to Hutchinson's treatise (1975), there were few comprehensive data sets for inorganic elements in freshwater aquatic organisms, and none of them were for Great Lakes biota. The most comprehensive data came from studies by Cowgill (1970, 1973*a*, 1973*b*, 1974*a*, 1974*b*), which Hutchinson cited extensively. Most of the data sets for the elemental composition of aquatic species cited in the literature of radiochemistry and radiobiology were for marine organisms. From a radiochemical perspective, the situation logically followed from the knowledge that the oceans were the main repositories for many of the nuclides, especially those from atmospheric fallout or discharged to rivers and estuaries and carried downstream to the oceans. The need for risk assessments for radionuclides to humans required data on food stuffs, and this caused an emphasis on those marine fishes and shellfishes harvested for human consumption. Thus, the early investigators of aquatic systems (*e.g.*, Vinogradov 1953; Bowen et al. 1971) focused on nuclide uptake by marine biota.

Concern for freshwater species has recently increased, but the motivation comes from the desire to understand the toxicology of the elements and their environmental effects and not from any interest in bioaccumulation processes. Bioaccumulation processes became a research priority with the discovery that several types of organic compounds became biomagnified as they cycled through ecosystems via foodwebs. These compounds were listed under the Great Lakes Water Quality Agreement of 1978 as "persistent toxic chemicals." A few inorganic elements belong to the category of persistent toxic chemicals: mercury, cadmium, and arsenic.

Pre-1970 data on bioaccumulation and biomagnification

Studies of marine biota have the produced most of the high-quality bioaccumulation data that are given in the tables and compilations of bioaccumulation and biomagnification factors found in the technical literature. Marine data are also among the oldest data, with many citations to two of the most widely accepted and used references: the volume by the United States National Academy of Sciences (1971), *Radioactivity in the Marine Environment* and the report by Chapman *et al.* (1977), *Concentration Factors of Chemical Elements in Edible Aquatic Organisms* (UCRL-50564). Table 2 summarizes the data of Chapman and coworkers.

TABLE 2
FRESHWATER BIOACCUMULATION FACTORS FOR VARIOUS ELEMENTS
(based on Chapman *et al.* (1977) — UCRL-50564)

Element	Plants	Invertebrates	Fishes
Sodium	200	30	30
Chromium	4000	2000	200
Manganese	10,000	40,000	20
Iron	5000	3000	300
Cobalt	1000	2000	500
Nickel	100	100	40
Zinc	4000	40,000	1000
Silver	200	3000	3000
Tungsten	30	30	1
Rubidium	1000	2000	2000
Strontium	500	700	40
Yttrium	10,000	1000	100
Zirconium	10,000	1000	100
Niobium	1000	100	30,000
Molybdenum	100	100	100
Phosphorus	100,000	100,000	100,000
Ruthenium	2000	2000	100
Rhodium	2000	2000	100
Antimony	—	2000	40
Tellurium	200	200	400
Iodine	100	20	4
Cesium	300	1000	1000
Barium	500	200	10
Rare earths (Ce, La, Nd, Pr)	10,000	1000	100
Neptunium	1000	300	10,000
Promethium	10,000	1000	200
Tritium	1	1	1

Notes: The factors are based on radionuclide uptake relative to the water. Bioaccumulation factors based on the consumption of plants and invertebrates, or on organisms found in benthic or sediment environments relative to the sediments are not given. Bioaccumulation factors are referred directly to water, not to the lower trophic level of an implied food chain. Therefore, to estimate biomagnification factors, one must take the ratio of the data in a given column to the ratio of the column which is at its left (the preceding column).

The Task Force emphasizes the following important caveats and commentaries accompanying the tables in *Radioactivity in the Marine Environment* (1971): (1) bioaccumulation factors typically have an *order of magnitude* uncertainty; (2) the tables do not consider differences between nearshore and open-ocean environments, surface waters and bottom waters or gradients of chemicals in the water column, nor the environmental gradients of salinity in estuaries; and (3) data do not account for different elemental sensitivities between life stages of organisms. The bioaccumulation factors in these tables carry an inherent uncertainty of 1000% or more, which may propagate errors when these factors are used in mathematical models of ecological uptake, transport, distribution, and species interactions.

The use of marine bioaccumulation data to develop inventories for radionuclides in freshwater biota of the Great Lakes radionuclides has severe limitations. Oceans differ from lakes in their hydrodynamic regimes, chemical compositions (especially with regard to salinity), species array, and food web dynamics. The bioaccumulation and biomagnification patterns for marine food webs tend to show a strong uptake of chemicals at the primary producer level (algae), further uptake in first-level consumers (most zooplankton in the form of

Euphausiads and calanoid copepods, and benthos in the form of molluscs and marine worms), and further uptake and accumulation in secondary consumers (fishes, birds, and sea mammals). Lakes often show a different pattern of uptake and accumulation at the first consumer level of zooplankton crustacea, protozoa, and insects.

Insects exhibit the most discrimination in their uptake and retention of chemicals from their food. Insects also dominate the invertebrate fauna of most lakes and rivers. Except for certain Lepidoptera and estuarine Corixidae, insects are very rare or absent from marine systems. Other benthic species express food preferences and discriminatory behaviors in bioaccumulation and retention of various elements and compounds. Hutchinson's (1995) cautions and comments on the vegetative preferences of freshwater gastropods are noted here.

The uptake, retention, and bioaccumulation of individual elements within tissues do not occur independently of one another. Elements move coherently with the uptake, retention, metabolism, and excretion of one element dependent on other elements. When an element's basic chemistry depends on or is affected by levels of sodium, calcium, chloride, carbonate, and sulfate ions, the biouptake processes will show major differences and gradients among freshwater, estuarine, brackish, and heavily saline or marine waters.

Calcium control paradigm

The Task Force examined data on the distribution of elements in both the environment and within biota for both radioactive and stable isotopes of many elements and noted their behavior in high- and low-calcium environments. These environments may be external (lake waters or lake sediments) or internal (cell contents or tissue distributions). Concentrations of calcium appear to correlate and interpret many of the observed situations and results. This applies to elements varying from barium and phosphorus to cerium and lanthanum. It appears that calcium *controls* the limnological behavior of many other elements, especially with regard to bioaccumulation and cycling through biotic communities and ecosystems. This led the Task Force to propose a *calcium control paradigm*.

Calcium within biota interacts with hormones, and because it usually exists in ionic form it can activate or interact with ionic channels. Other elements that can activate ionic channels are sodium, potassium, magnesium, and chlorine as the chloride ion.

The impoverished sodium content of fresh waters precludes this element from exerting the kind of widespread control needed because organism adaptations for low sodium have emphasized cell osmotic pressure. The single proton charge limits sodium's impact on cell systems relative to the calcium ion's divalent charge. Sodium also interacts with very few biomolecules. In marine systems, the environmental excess of sodium induced evolutionary changes *to reject the element* from many cellular systems to maintain cellular osmotic pressure.

Potassium occurs in relatively limited quantities in the natural lithosphere, although about 40% of the radioactivity of biota comes from the naturally occurring radionuclide, ^{40}K. Any consideration of potassium should be undertaken only after elimination of other control element possibilities.

The chloride ion is the major negative ion but has little effect on cellular pH. The other major negative ions (hydroxide, bicarbonate, and phosphate), through complex equilibrium relationships in cell fluids, mainly affect cell pH. Chloride serves mainly to provide the appropriate electroneutrality of biological systems. In certain marine teleost fishes, the chloride ion works with the bicarbonate ion to maintain osmotic pressure.

Magnesium plays an important biochemical role, but again, its limited geochemical occurrence makes it a lesser candidate for exerting widespread biological controls. Magnesium is a cofactor in metabolic reactions rather than through the operation of hormonal cascades. Magnesium also forms many covalent organo-magnesium compounds (*e.g.*, the classical Grignard reagents) more than does calcium.

Calcium is the most widespread inorganic ion in the lithosphere. It, along with magnesium, sets the "hardness" of the freshwater environment through its presence as a carbonate and prescribes the "alkalinity" of the marine environment through its presence as a carbonate or sulfate. Its widespread presence in the marine environment provides the major cation to neutralize a divalent sulfate anion.

The early history of water-quality criteria noted that "hard" waters often offered organisms greater protection from the toxicity of heavy metals than soft waters. Calcium blocks the toxicity of nickel, copper, zinc, iron, and cadmium by increasing the pH of the medium to the point where these metals remain in precipitated form as sulfate, carbonate, or other salts. Calcium ion provides the major buffering capacity of lakes against acidic precipitation. Calcium also blocks the receptor sites that are easily attacked by these heavy metals in a manner comparable with the way potassium iodide tablets block sites in the thyroid against radioactive iodine — through saturation by a competitive atom, a "mass action law" effect. Thus, calcium outside and calcium inside the cellular environment protect biota.

In the early history of limnology, waters were classified based on their hardness. As Balment and Henderson (1987) noted, the number of species often found in freshwater systems and their biomass correlate with the calcium levels of the environment, providing selected adverse factors are missing.

Also, the uptake of a radioactive form of an element often depends on the existing accumulation of a nonradioactive form of the same element (known as the "carrier" effect), a common example being radionuclides of iodine. The consumption of potassium iodide by humans to protect against uptake of radioactive iodine is a standard practice following a nuclear incident or upon exposure to radionuclides as radioiodine is a thyroid carcinogen.

Based on the previous discussion, the following elements would likely behave differently in their uptake by freshwater lake biota relative to marine biota: iodine, bromine, silver, zinc, cesium, rubidium, barium, strontium, manganese, cobalt, iron, molybdenum, sulfur, selenium, and lanthanum. Complicating the picture is the large-scale chemical pollution of the oceans and the Great Lakes. The documentation from nuclear power plants on the presumed behavior of nuclides following discharge into the environment has usually ignored the existing pollution problems and used estimates based on mathematical models with the bioaccumulation factors from tables derived from studies of the marine organisms.

Nuclides "in" and nuclides "on" organisms

An interesting problem in studying bioaccumulation of radionuclides is to distinguish between nuclides "on" the organism and nuclides "in" the organism. *On* means on the surface; *in* means inside the organism, either in tissue as a free nuclide or bound to some protein or receptor. Nuclides on an organism might be"loose" and easily washed off, or they may be bound to the surface because of the secretion of some adhesive type of biological material. The following are some specific examples.

(1) Silicon. This element is a fundamental component of the frustules of diatoms, and remains as a fossil record of the species. Many diatoms are epiphytes on other substrates, including macrophytes and stalked algal species. Very high silica analyses may represent the surface contamination of silica by diatom frustules.

(2) Calcium. Calcium usually occurs as the carbonate in freshwater systems. As freshwater organisms respire, they excrete carbon dioxide into the aquatic medium. The carbon dioxide then undergoes the chemical reactions of the "carbonate equilibrium" to produce carbonate ion, which ties up calcium ion. Some calcium carbonate precipitates on the surface of vegetation. Additional chemical changes occur because of shifts in the carbon dioxide – carbonate equilibria to make carbon available for photosynthesis thereby reducing environmental levels of carbonate and releasing calcium to the aquatic medium.

(3) Iron and manganese. Both elements form insoluble, gelatinous, and crystalline oxide complexes. Under appropriate conditions of pH, these oxides will precipitate on surfaces. The carbon dioxide – carbonate equilibria are major determinants of the pH of a waterbody, and thus organismal respiration and plant photosynthesis may locally affect the pH, leading to iron and manganese oxide precipitation and deposition on plant tissue or other substrates (including possibly a floating gelatinous precipitate in the water column). Many analyses showing high levels of these two elements, despite their metabolic roles in organisms, reflect surface contamination of the sample by precipitated oxides.

(4) Titanium. This element can coat surfaces as the oxide form and is not easily incorporated into tissues, although titanium citrate has a limited solubility. Compounds of titanium can pass through the alimentary canal of animals if in or on the food. Only a material balance on the excreted materials of the species will enable one to tell if titanium has been retained in the organism.

The uptake of some elements is a two-step process: first a surface contact process that often can be described in terms of the area of contact or exposure, then a cell uptake process, which may require a diffusional mechanism, an active transport mechanism, or a chemical reaction. For some plants, the first process might mean entry through stomata or roots. For other species it may entail transport across a biological membrane or into a vacuole directly from the external medium. When nuclides on the organism are a surface contamination of the sample, the chemical analyses overestimate the uptake. If the nuclide levels on an organism quantify fallout of the nuclide, the information has separate value and importance. Nuclear power plants monitor the radiation levels at nearby farms, agricultural, and forestry tracts to assure that their nuclide emissions do not contaminate food supplies. The problem described has raised challenges to the quality of considerable data on nuclide bioaccumulation and biomonitoring in terrestrial systems.

Biomagnification of metals attached to organic molecules

According to DeFilippis and Pallaghy (1994), heavy metals do not typically follow the biomagnification patterns and processes of organic chemicals cycling through aquatic food webs. Thus for nuclides of heavy metals, algae would typically have greater concentrations of the nuclides than zooplankton predators and fish. This does not appear to be the case for metals attached to organic molecules, notably methylated compounds of mercury, tin, arsenic, lead, and cadmium. Therefore, radionuclides that can attach to hydrophobic molecules, and thus become lipid soluble, will likely biomagnify as they cycle through aquatic food webs.

Other biomagnification processes are not exclusively the result of predator–prey (consumer–food) interactions. Mercury in the water or sediment may equally have provided the sources of mercury observed in the tissue of organisms. Further complicating the situation is the possibility that a previously biologically unavailable source of a nuclide became available in the digestive system of an organism through methylation reactions *in vivo*.

The biochemistry of the processes by which metals attach to organic molecules, other than an ion-exchange process (simple binding replacement of H^+), usually involves the presence of one or more of the following: Fe, Mn, Co, pyridoxine, NADP, ATP, or a sulfhydryl group. Their presence suggests interactions among elements. Since the processes are often microbially mediated in the environment rather than in the tissues of plants and animals, the half-life of the radionuclides is the major determining radiological factor, and appropriate populations of microorganisms and available food supplies are the major determining ecological factors. These processes will occur if radiation does not have a sterilizing effect.

Use of nuisance species for bioaccumulation studies

If nuisance species dominate or represent major components of the biotic assemblage of a region, then it would seem only natural to study their elemental composition and cycling as part of the intensive understanding of their metabolism, physiology, ecology, and behavior. This approach has found favor in selected European studies and recently in selected studies in North America. Particular useful are the recent studies of the Asiatic clam, *Corbicula flumatilis*, in San Francisco Bay, connecting tributaries, and lakes. *Corbicula* has spread rapidly eastward through North America since its introduction into California some 20 years ago and now occurs in several tributaries that feed the Great Lakes.

The sea lamprey, *Petromyzon marinus*, which entered the Great Lakes through the Saint Lawrence Seaway, has devastated the lake trout, *Salvelinus namaycush*. The lamprey has been the focus of an intensive binational effort through the Great Lakes Fisheries Commission to develop selected chemicals for its destruction, yet the Task Force finds it somewhat surprising that its elemental composition has gone unreported.

At one time a well known statement was "Lake Erie was dying." The alga, *Cladophora glomerata*, formed huge mats of floating and decaying vegetation that washed up on the beaches of Ontario and New York. *Cladophora* has received considerable attention for its elemental bioaccumulation because it is a known concentrator or accumulator of heavy metals, but the physiological factors that might enhance biouptake and the use of *Cladophora* as a mechanism for detoxification of elements and radionuclides in the Great Lakes were never really effectively explored. Because of its rapid growth under nuisance conditions, Whitton (1984) has shown that the young tips can be used to differentiate between recently accumulated pollution and older or long-term pollution. The species has a relatively high thermal tolerance so that its peak growth periods are late spring through summer and early fall. Further, *Cladophora* seems to have a special obligate requirement for sodium. Levels of sodium have increased in the Great Lakes because of human activities, but appropriate correlations with *Cladophora* were not reported in the literature. Rather, the emphasis on nutrient control (mainly nitrogen and phosphorus), sought to remove the basic building blocks from pollution sources for excessive algal growth. That strategy has worked well, and it no longer appears that *Cladophora glomerata* has nuisance abundance in the Great Lakes. Nevertheless, the species does exist and coexist with the biotic assemblages and could still serve as a sentinel in tracking the cycling of radionuclides if such work were included in existing research and monitoring studies.

Another nuisance species introduced into the Great Lakes through is the zebra mussel, *Dresseina polymorpha*. This species has caused a major shift in the plankton. The mussel's radionuclide content will reflect not only direct uptake from the aquatic environment but nuclides from the filtered phytoplankton species which it consumes. The species has received some attention for its elemental bioaccumulation in European studies, but the Task Force is not aware of comparable studies for North America.

BIOUPTAKE DIRECTLY FROM ATMOSPHERIC DEPOSITION

Many organisms can draw chemical substances directly from the atmosphere. Since the work of the Task Force emphasizes the Great Lakes, the concern will focus on plants, fungi, and related species that can provide insight into cycling of elements through biocompartments of the Great Lakes ecosystems.

The direct utilization of atmospheric components usually depends on the state of matter of the chemical species. Chemical substances in the gaseous or vapor phase can penetrate the air breathing apparatus most easily. If those chemicals readily dissolve in water, the contact of the substances with water on an organism may further facilitate penetration of the organism. Aerosols and particulate matter have greatest difficulty, usually requiring some preliminary processes simply to make the substances available to the organism.

Two important groups of organisms that can bioaccumulate materials from the atmosphere are lichens and mosses. These organisms have played a very important role in studying the deposition of atmospheric substances, both radioactive and nonradioactive, especially those chemicals in particulate and aerosol form. Lichens are emphasized in the materials. It is simply that lichens have a better studied and understood ecological context with respect to the uptake of radionuclides and their transfer to other species.

For chemicals in particulate form, lichens generally accumulate substances by a two-stage process: a contact or binding stage, which is well described thermodynamically as an ion-exchange process, followed by a tissue absorption and retention phase. The second phase typically uses a carrier molecule.

From a kinetic perspective, the relative time scales of the two stages are important in quantifying rates of bioaccumulation. Yet, the experimental and statistical methods used to study bioaccumulation processes rarely address the differences in time scales for the tissue absorption and retention steps.

For many of ecosystems, lichens are the major food source for selected mammals, notably deer and caribou. Further, sometimes lichens are the *only or major* food source for these mammals. Mammalian consumption of lichens is direct route of transfer and exposure of radionuclides regardless of how radionuclides may occur in the lichens. The mammalian digestive system of these species does not differentiate between the binding modes in the lichens because the system breaks down the lichen cell matter and makes nuclide absorption readily available in the mammal's stomach and intestines.

The ion exchange nature of the binding is easily confirmed statistically. A simple model of ion exchange allows for *equal and independent* competition of ions for sites among possible binding nuclides. The particulate matter and aerosol forms of the metallic element radionuclides of the following elements are usually oxides: Na, K, Rb, Cs, Ag, Be, Ca, Ba, Sr, Ra, Ce, La, Fe, and Ru. Contact with water releases these metals as cations which can exchange for replaceable (*sensu*: ionizable) hydrogens on the lichen. The univalent Na, K, Cs, and Ag cations would each replace a single ionizable hydrogen; the divalent Be, Ca, Ba, Sr, and Ra would replace two ionizable hydrogens; and the trivalent Ce and La would replace three ionizable hydrogens.

Since the geochemical ratio of ^{90}Sr/^{137}Cs is important in evaluating depositional data of radionuclides, the behavior of an appropriate geochemical ratio with respect to bioaccumulation in lichens is derivable from the ion-exchange model. The model would suggest a strong correlation of the geochemical ratio for the two nuclides in atmospheric particulate matter and that bound to the lichens. Small deviations of the geochemical ratio reflect the reality that the binding process is not 100% efficient and that any two-stage processes of absorption and tissue retention would tend to favor strontium over cesium in most species. The two-stage model basically assumes that the uptake process is divided into separate binding and uptake steps. The first stage is a Langmuir isotherm equilibrium process for an ion in the atmosphere attaching to a suitable site on the surface of a plant. Once this binding has occurred, the plant may secrete carrier molecules that can bind to the ion and transport into the cell, or openings in the cell (stomata) allow the movement by diffusion of the ion into the tissue, or some similar process. The rates at which the two stages occur differs, and this difference in rates determines the extent to which the two-stage model correlates the available kinetic data. If the first stage is very fast, the slower second stage will determine the uptake. Materials still in the first stage could theoretically be washed off the surface binding sites. If both stages have process rates within the same numerical order of magnitude, both surface leaching and tissue uptake will be observed. If the second stage is much faster than the first, only tissue-based biouptake will be observed. Similarly, radionuclides of Br, Cl, I, Al, Mn, Tc, and Ru, which exist as anions, would require a different mechanism of tissue uptake. If a lichen had anionic sites, a second model of ion exchange would should behave similarly for anions as the previously described one for cations. Again, such a model would also permit assessment of the statistical behavior of appropriate geochemical ratios.

Several elements occur environmentally in several different forms, making a model based on ion exchange processes inadequate. The two main examples are sulfur and nitrogen. Forms of sulfur require special treatment because they interact with metallic elements. The chemistry of nitrogen is also complex, but if the form of nitrogen present in the environment is inorganic, it can often be predicted from a knowledge of pH and oxygen levels through a consideration of the ammonia–nitrate–nitrite equilibria. Nitrogen, in ammonia, can bind ionically to lichens in the same manner as cations, but nitrite, and nitrate are anionic and would not bind to the ion exchange sites in lichens that are designed for cations.

Nitrogen is also the elemental basis for proteins. Radionuclides of nitrogen are not a problem with respect to the Great Lakes, although the element is intricately tied to the problems of acid precipitation and eutrophication. Nitrogen atoms are a source of cosmogenically derived radionuclides, but these nuclides have chemistries that can be treated on their own.

Several elements can exist in ionic and covalent forms. The covalent bond offers additional mechanisms of biouptake, including the solubilization in lipid materials and transport across certain types of hydrophobic membranes. Such elements include Hg, Cd, Sn, Te, Se, As, P, and, again, various forms of nitrogen.

The Task Force did not access data on lichens in the Great Lakes region. Most of the available lichen data refer to restricted and extreme climate and geographical zones: Arctic, high altitude, tropical regions in the vicinities of volcanic fumaroles, and selected forest regimes. Since lichens often dominate the flora in the area in which they reside, one need not consider many of the implications of species diversity on the reported values of elemental composition and bioaccumulation factors.

Table 3 describes a tropical lichen in the vicinity of a volcanic fumarole in Hawaii. The purpose of O'Connor's study (1979) was to track mercury from the volcanic emissions and its long range transport effects on nearby and distant vegetation. The usefulness of the data is in its coverage of a large number of elements. Radionuclide deposition data for the region for ^{90}Sr and ^{137}Cs are available, and the data array can be used to compare lichen behavior for bioaccumulation in a more generic manner.

Table 4 presents data for lichens and mosses which have been studied for their content of several cosmogenically produced radionuclides and two groups of radioactive decay series nuclides from transuranics. These data from Jenkins *et al.* (1972) are from one of the classic studies on natural background levels of radiation.

TABLE 3
CHEMICAL COMPOSITION OF A TROPICAL GROUND LICHEN (*Cladonia skottsbergii*)
(from O'Connor 1979)

Sample description	**Elements (ppm)**										
	K	**Cl**	**Ag**	**Mg**	**Ca**	**Ba**	**Sr**				
Lichen (0.1 km from volcano)	24,666	350*	<0.44	43,033	11,200	140	457				
Lichen (2.8 km from volcano)	28,333	350*	0.95*	10,766	13,333	157	553				
	Ti	**V**	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Zn**	**Zr**	**Nb**	**Mo**
Lichen (0.1 km from volcano)	9166	183	377	927	46,000	3.7*	180	400	97	<9.3	25.3
Lichen (2.8 km from volcano)	8567	170	357	887	41,000	<0.92	177	270*	88	20*	10.2
	Al	**B**	**Ga**	**Y**	**Sc**	**P**	**Ge**	**Pb**	**S**	**Se**	
Lichen (0.1 km volcano)	43,000	38.6	3.1*	17	26	4333	1.4	44.1	233	0.77	
Lichen (2.8 km from volcano)	43,667	45.3	2.1*	22	24	3000*	1.2	55.0	233	1.6	

Notes: Results are average of three samples except those marked with an asterisk (*), which are based on either one or two samples.

TABLE 3 (continued)
CHEMICAL COMPOSITION OF A TROPICAL GROUND LICHEN (*Cladonia skottsbergii*)
(from O'Connor 1979)

Sample description	**Geochemical ratios**										
	Mg/Ca	**Ba/Ca**	**Sr/Ca**	**Ca/Ti**	**V/Ti**	**Zr/Ti**	**V/Nb**	**Fe/Mn**	**Co/Fe**	**Ni/Fe**	**Co/Zn**
Lichen (0.1 km from volcano)	3.84	0.0125	0.0408	1.22	0.0199	0.0105	—	49.6	0.00008	0.0039	0.00925
Lichen (2.8 km from volcano)	0.807	0.0117	0.0415	1.56	0.0198	0.0102	8.5	46.2	—	—	—
Earth's crust	0.561	0.001	0.009	7.37	0.0237	0.0289	6.8	59	0.00044	0.0013	0.333
	Enrichment factors										
Lichen (0.1 km from volcano)/earth's crust	6.84	12.5	4.53	0.166	0.839	0.363	—	0.841	1.82	3.0	0.0278
Lichen (2.8 km from volcano)/earth's crust	1.44	11.7	4.61	0.212	0.835	0.363	1.25	0.783	—	—	—

Notes: Enrichment factors are calculated relative to crustal material. Thus, values greater than 1 for enrichment factors means that organisms retain the elements to a greater extent than is found in natural soils or crustal materials. This suggests that bioaccumulation is important for barium, calcium, strontium, magnesium, iron, manganese, vanadium, cobalt, and nickel and of limited potential for titanium, zirconium, and zinc.

Sample description	**Geochemical ratios**								
	Cr/Mo	**Al/B**	**Ga/Al**	**Y/Ti**	**Sc/Ti**	**P/Ti**	**Pb/Ti**	**Ge/Ti**	**Se/S**
Lichen (0.1 km from volcano)	14.9	1114	0.00007	0.00185	0.00284	0.101	0.0010	0.00003	0.0033
Lichen (2.8 km from volcano)	35.3	963	0.00005	0.00257	0.00280	0.064	0.0012	0.00003	0.0069
Earth's crust	66.6	8240	0.0018	0.0058	0.0039	0.184	0.0022	0.00263	0.00019
	Enrichment factors								
Lichen (0.1 km from volcano)/earth's crust	0.224	0.135	0.0388	0.319	0.728	0.549	0.454	0.0114	17.4
Lichen (2.8 km from volcano)/earth's crust	0.530	0.117	0.0278	0.443	0.483	0.348	0.545	0.0114	36.3

Notes: Enrichment factors are calculated relative to crustal material. Thus, values greater than 1 for enrichment factors means that organisms retain the elements to a greater extent than is found in natural soils or crustal materials. This suggests that bioaccumulation is important for molybdenum, selenium, and sulfur and of limited potential for scandium, yttrium, phosphorus, and lead.

TABLE 4
NATURALLY OCCURRING RADIONUCLIDES IN LICHENS AND MOSSES FROM A FOREST IN WASHINGTON STATE, 1966–1967
(from Jenkins *et al.* 1972)

Species	Location	Radionuclides (pCi/kg wet weight)					
		^{7}Be	^{22}Na	^{40}K	^{208}Tl–^{232}Th	^{214}Bi–^{226}Ra	$\sum$TU
Lobaria oregana (lichen)							
Hoh River rain forest	Closed canopy	8400	—	2900	4.1	18	22.1
Queets rain forest	Closed canopy	20,000	5.2	3200	9.3	48	57.3
Quinalt rain forest	Closed canopy	7900	2.4	2000	7.6	17	24.6
Selaginella serpens (moss)							
Hoh River rain forest	Closed canopy	330	0.6	1700	—	4.5	5.5
Queets rain forest	Closed canopy	3700	2.2	1300	6.0	16	22
Quinalt rain forest	Closed canopy	3900	1.5	2900	15	110	125
Atmosphere	over Richland, Washington						
Air, November 1966		4.7	0.0003		0.0004	0.0008	0.0012
Air, March 1967		6.1	0.0013		0.003	0.002	0.005
Air, January 1968		5.4	0.0006		0.001	0.004	0.005
Air, December 1967	over Olympic Peninsula, Washington	5.8	0.0005		0.0006	0.0004	0.001
Rainfall, November 15, 1967		12.0	—		0.0018	0.0045	0.0063
Rainfall, March 21, 1967		26.0	0.0077		—	—	
Rainfall, January 11, 1968		30.0	0.0084		—	—	

Notes: (1) Sampling in the Queets rain forest and Quinalt rain forest during heavy rainfall on March 22–23, 1967

(2) Symbols: $\sum$TU (total transuranic decay series nuclides), obtained by summing the nuclide information in the two columns which provide information on the decay series nuclides (Th and Ra).

(3) These data are based on wet weight, and would not meet the requirements in Text Box 4. However, these data have historical importance for their inclusion of two cosmogenically formed radionuclides, ^{7}Be and ^{22}Na. Further, the data set reflected the effects of rainfall on the forested system, and the documentation of such effects is rare in nuclear literature.

TABLE 4 (continued)
NATURALLY OCCURRING RADIONUCLIDES IN LICHENS AND MOSSES FROM A FOREST IN WASHINGTON STATE, 1966–1967 (from Jenkins *et al.* 1972)

Species or substrate	Geochemical ratio				
	^{7}Be/^{22}Na	^{7}Be/^{40}K	^{22}Na/^{40}K	$\sum$TU/^{22}Na	$\sum$TU/^{40}K
Air	9490	—	—	4.54	—
Rainfall	3474	—	—	—	—
Lichens					
Hoh River rain forest	—	2.89	—	—	0.0076
Queets and Quinault rain forests	3569	5.10	0.0015	10.6	0.0151
Moss					
Hoh River rain forest	500	0.194	0.00039	9.17	0.0032
Queetc and Quinault rain forests	2141	2.10	0.00098	46.7	0.030

	Bioaccumulation factor							
	^{7}Be	^{22}Na	^{40}K	$\sum$TU	^{7}Be/^{22}Na	^{7}Be/^{40}K	$\sum$TU/^{22}Na	$\sum$TU/^{40}K
Lichens/air								
Hoh River rain forest	1556	—	—	6697	—	—	—	—
Queete and Quinault rain forests	3417	2923	—	6120	1.17	—	2.09	—
Lichens/rainfall								
Queete and Quinault rain forests	537	494	—	—	1.08	—	—	—
Moss/air								
Hoh River rain froest	61.1	857	—	1618	0.072	—	1.89	—
Queete and Quinault rain forests	704	2642	—	21,617	0.266	—	8.18	—
Moss/rainfall								
Queete and Quinault rain forests	146	240	—	—	0.608	—	—	—
Air/rainfall	0.25	0.42			0.59			
Lichens/mosses (air)								
Hoh River rain forest	25.5			4.13				
Queete and Quinault rain forests	4.9	1.1		0.28	16.3		0.26	
Lichens/mosses (rainfall)								
Queete and Quinault rain forests	3.7	2.1			1.8			

Radionuclide data from Arctic ecosystems

The deposition of radionuclides of cesium from the atmosphere in the northern hemisphere favors the northernmost latitudes. Some of the ecological systems in these latitudes are rather limited because of the climate conditions. An important system in these regions is the lichen – reindeer/caribou – human food chain. Lichens, a plant–fungus symbiont, can readily accumulate metallic elements because some of the plant material behaves like an ion-exchange resin. Metallic elements in particulate matter and aerosol materials of atmospheric fallout will collect on the lichens and can displace bound hydrogen atoms. Cesium ions are positive and univalent and readily displace hydrogen in ion-exchange systems. Thus, the chemical behavior of radionuclides of cesium, as in ^{137}Cs, assures that, if it contacts lichens, it will be incorporated into the tissue through this ion-exchange mechanism. Reindeer and caribou eat the lichens, and these mammals are in turn the food of resident populations. This food chain pathway to man of the radionuclides of cesium has been studied in various Arctic regions, including northern Canada, Alaska, Denmark and the Faroë Islands, Sweden, Norway, Finland, and Commonwealth of Independent States (formerly Russia). Table 5 lists the concentrations of cesium and strontium in Arctic biota.

The studies of Arctic species and radioactive cesium loadings go back to 1969. UNSCEAR (1977) reported on the work through the early and middle 1970s. Following the accident at the nuclear power plant at Chernobyl (Ukraine) in 1986, researchers and Government agencies undertook new studies of atmospheric deposition in the Arctic. Many of the results of these studies appeared in 1995 in a special symposium on pollution in the Arctic (Volume 160/161, *Science of the Total Environment*). The research findings updated important information on ^{137}Cs levels in Arctic species, including some species not previously considered. (Arctic Monitoring and Assessment Program, 1997.)

TABLE 5
RADIONUCLIDES IN ARCTIC BIOTA (LICHENS AND MOSSES)

Study: Nifontova (1995)
Radionuclides in lichens and mosses from Yamal Peninsula (Russia), an Arctic Region

Nuclide activity (Bq/kg)

Species	**^{90}Sr**	**^{137}Cs**	**^{90}Sr/^{137}Cs**	**Species**	**^{90}Sr**	**^{137}Cs**	**^{90}Sr/^{137}Cs**
Lichens:				Mosses:			
Cladina rangiferina	40 ± 10	315 ± 30	0.12	*Dicranum spadiceum*	160 ± 40	170 ± 10	0.94
C. sylvatica	50 ± 10	280 ± 30	0.18	*D. elongatum*	110 ± 15	270 ± 20	0.40
C. stellaris	40 ± 10	150 ± 30	0.27	*Hylocomium splendens*	130 ± 10	120 ± 10	1.08
C. arubscula	70 ± 10	220 ± 30	0.32	*Polytrichum commune*	150 ± 20	340 ± 60	0.44
C. mitis	50 ± 10	320 ± 30	0.16	*P. strictum*	50 ± 10	150 ± 10	0.33
C. uticalis	—	350 ± 50		*P. gracilis*	90 ± 20	80 ± 20	1.13
Cladonia elongata	150 ± 20	160 ± 20	0.94	*Rhacomitrium lanuginosum*	160 ± 30	410 ± 10	0.39
C. amaurocreaea	110 ± 30	335 ± 50	0.31	*Sphagnum riparium*	110 ± 20	300 ± 30	0.37
Cetraria chrsantha	30 ± 5	180 ± 30	0.17	*S. rubellum*	120 ± 20	220 ± 20	0.55
C. hipatison	90 ± 20	140 ± 20	0.64	*S. squarrosum*	30 ± 10	120 ± 20	0.25
C. islandica	40 ± 5	280 ± 90	0.14	*S. wulfianum*	60 ± 10	80 ± 20	0.75
C. laevigata	30 ± 10	50 ± 10	0.60	*S.majus*	70 ± 10	90 ± 10	0.78
Thamnolia vermicularis	120 ± 20	140 ± 10	0.86	*S. fuscum*	130 ± 10	330 ± 50	0.39
Sterocaulon tomentosum	60 ± 10	40 ± 10	1.5	*S. balticum*	150 ± 10	360 ± 90	0.42
				S. lenense	120 ± 30	260 ± 50	0.46

TABLE 5 (continued)
ELEMENTAL ANALYSES (INCLUDING RADIONUCLIDES) OF ARCTIC BIOTA

Study: Nifontova (1995)

Average activity of nuclides (Bq/kg)	^{90}Sr	^{137}Cs
Cladina (5 spp.)	50	257
Cladonia (2 spp.)	130	247.5
Certraria (4 spp.)	54.8	162.5
Lichens (13 spp.)	64.6	200.8
Lichens (14 spp.)		211.4

Average activity of nuclides (Bq/kg)	^{90}Sr	^{137}Cs
Dicranium (2 spp.)	135	220
Polytrichum (3 spp.)	96.7	90
Sphagnum (8 spp.)	98.8	220
Mosses (15 spp.)	109.3	220

Geochemical ratios for ^{90}Sr/^{137}Cs:

Nuclear fallout	0.625
All lichens	0.478
All mosses	0.579
Lichens + mosses	0.532

Enrichment factors (EF) for plants subject to nuclear fallout:

(EF) lichens/fallout	0.765
(EF) mosses/fallout	0.926
(EF) (lichens + mosses)/fallout	0.851

Notes: "R" is the geochemical ratio based on the activity measurements indicated in the columns to the left. The 13 lichens are those for which there are both Sr and Cs data. The 14 lichens are for the entire collection of lichens. The geochemical ratios are based on 13 lichens and 15 mosses. The combined geochemical ratio for lichens and mosses is based on 28 plant species. "EF" is the enrichment factor, which is the ratio of geochemical ratios of the two indicated nuclides relative to their respective media or substrates (thus, EF for lichens/fallout is the ratio of the geochemical ratios for each nuclide in lichens to the geochemical ratio of the two nuclides in fallout).

BIOACCUMULATION IN SOIL MICROFLORA

There are several kinds of organisms important in the cycling of elements through ecosystems that can accumulate elements from air and soils. The soil layer microflora, especially the fungi, are very important, but this group of organisms is often overlooked in understanding the movement of elements through ecosystems. Because some fungi are edible (*e.g.*, selected species of mushrooms), the interest in accumulated elements has applicability to risk assessment to ecosystems and humans. Further, the fungi provide a comparison group to the lichens and mosses, which have been previously discussed. However, data sets on bioaccumulation of elements were available mainly for European species not North American species.

BIOLOGICAL PRODUCTIVITY

Because of the importance of biological productivity measurements in an discussion of bioaccumulation and biomagnification of various nuclides, this section begins with a short discussion of the subject and a review of the available data on biological productivity for various locations within the Great Lakes Basin. A fundamental research problem in biology is how biological systems sustain themselves. Scientists want to quantify those sustaining properties, the most of important of which is the ability to produce new protoplasm to replace the aging, injured, diseased, dead, or lost (removed) protoplasm while assuring that the new protoplasm maintains the speciation and distribution to sustain the different kinds (diversity) of species that form the biological system. The production of protoplasm in biological assemblages and ecosystems is *biological productivity*. This inventory of radionuclide is the product of two terms: the radionuclide activity in a biological compartment and the mass of that compartment. The term usually known to the Task Force is the radionuclide activity for some medium, substrate, or even compartment. Because the second term in the product is usually absent, the available data can only show the relative distribution of radionuclides within compartments but no overall *absolute* estimate of their presence.

The "metabolism of lakes"

Limnologists desired a unifying concept to analyze the nutrient-induced excess or luxuriant biomass productivity of lakes (eutrophication). Wilhelm Rhode convened a seminar on the subject of "lake metabolism" (*Ergebniesse der Limnologie*, 1979) in which he compared a lake to a living organism with a "metabolism" that can be studied much as the physicians study the metabolism of human patients.

Traditionally, the measurement of organismal metabolism tracks the oxygen supplied to the organism and used and the carbon dioxide returned. During periods of photosynthesis, plants partially reverse the process and consume carbon dioxide and return oxygen. However, photosynthesis rarely fixes all of the carbon dioxide produced through respiration and metabolism, and thus one typically observes diminished rates of carbon dioxide production during periods of photosynthetic activity relative to periods of nonphotosynthetic activity, rather than reversal of the direction of the metabolic measurements.

Since only plants and some photosynthetic protozoans can carry out photosynthesis in freshwater systems, productivity in consumer organisms moves in the same direction as metabolism; oxygen uptake and carbon dioxide release. Further, if there is organismal death or tissue destruction, the bacterial respiration release nutrients back to the pool, producing large amounts of carbon dioxide and creating oxygen deficits.

Whole-lake productivity sums the productivity of each biological compartment. Whole-lake metabolism sums the metabolism of each biological compartment. Depending on how one views the system, a biological compartment may consist of the population of a species or the population of a biological assemblage of species or the biomasses of the species that perform some collective ecosystem task of nutrient and energy cycling. Limnologists can usually measure some components of productivity and some components of metabolism, but not all of the components of each, and not always both components for a single biotic compartment. Therefore, limnologists seek predictive relationships between various kinds of productivity and metabolism data to obtain the whole-lake productivity and whole-lake metabolism as well as the productivity and metabolism of critical biotic parts of the lake ecosystem. Because of the complexity of many of these relationships, the prediction of the productivity components, especially in terms of speciation, is one of the great research problems guiding limnological investigation, and the motivation for the development of sensitive and widely applicable methods to measure the more difficult secondary productivity.

Because eutrophication complicates the relationships between metabolism and productivity, the Task Force cautions against extrapolating metabolic and productivity data from low nutrient systems to eutrophic systems, or even from one eutrophic system to another.

Direct measurements of the mass of an element in a biological compartment tend to be more difficult than direct measurements of the activity of a radionuclide, but the quantification of a radionuclide for purposes of establishing an inventory can be overwhelming. The difficulties *begin* with sampling of biological substrates, quality assurance on analytical methods and instrumentation, overcoming limitations at the "lower level of detection," the number of analyses required for a reasonable application of statistics, and the design of the protocols for the studies to obtain the nuclide data for an inventory.

Many of the problems mentioned in the previous paragraph require an iterative approach for their solution. Careful analyses and questioning enable refinement of sampling and analytical procedures. A consideration that strongly impacts the process of obtaining suitable data and calculating the inventories for nuclides and that presents a special barrier is that one must really know in advance how various chemical elements in the biological substrates in a given compartment relate or interact with each other as well as how different biological compartments relate and interact with each other. Such information is rarely known in advance of a study, but itself is gleaned from the work as a study progresses. That means one needs to begin with some rather educated guesses to minimize unproductive field and laboratory work.

Radioactivity adds to the problems encountered in an overall understanding of the processes of element uptake and cycling and associated observed phenomena in the following ways.

(1) A radionuclide is an unstable version of the element. Its radioactive decay may produce a nuclide(s) of a new element(s). Although the original element may have been incorporated into biological tissues, the daughter nuclides of different elements in those tissues invoke an entirely new set of chemical processes with different metabolic and physiological responses by the organism, introduce new element toxicity concerns, and require that the organism to use a different suite of detoxification and depuration mechanisms to protect itself. How the transport processes associated with a daughter nuclide will impact the transport processes of the parent nuclide is also of concern.

(2) Radioactivity adds its own toxicity to any chemical toxicity of the nuclide. Even non-metabolically active nuclides may cause physiological harm through radiation–matter interactions: internal irradiation. Further, internal irradiation continues as long as the tissues retain the nuclides.

The "trophic cascade" concept

Biological productivity in the Great Lakes is a complicated set of processes and include the strategies of predation of the indigenous biota, the invasion of species from foreign habitats, shifts in habitat structure within the Great Lakes and the presence of many different kinds of pollutants, both radioactive and chemical. As used by Carpenter and Kitchell (1993), this model is one of a "trophic cascade" that attempts to "bridge ecosystem and population ecology." The former discipline tends to emphasize large scale processes of nutrient and energy cycling through complex compartments while the latter discipline examines more microscale processes within species or biotic assemblages without necessarily reflecting ecosystem organization. One begins with simple food-web relationships of primary productivity and detrital production, adds the consumption of these resources by invertebrates in plankton and benthos and the higher trophic level consumption of these resources by fishes, reptiles, birds, and mammals, as well as the feedback pathways which regenerate previously considered compartments.

A "cascade" implies some kind of "domino effect" or "amplification phenomenon" in a system. Each isolatable or definable food chain or food web subunit could manifest a possible cascade. By examining the relative rates of productivity of different elements of the food web as influenced by the population-level strategies of food choices, prey selection and acquisition, selective toxicity, and differential mortality of abiotic and external factors (*e.g.*, harvesting, cropping, chemical toxicity, stochastic effects of climate change), one seeks a description of how the entire food web functions. If the effects of changes in one element of a food web propagate through the food web to produce a rapid and, sometimes, highly unpredictable realignment of the food web structure, such an effect has "cascaded."

The occurrence of a cascade requires that the food web have, as a minimum, either a point of unusual vulnerability or the interactions of various components of the food web during energy and chemical cycling have some basic *nonlinear* (*sensu:* mathematical rather than biological) property to permit an amplification of some effect through an entire system. A point of vulnerability usually has the characteristic of a positive feedback element above some threshold system value. For example, some pathway becomes *saturated* and loses its stabilizing contribution to the food web, or the pathway becomes *damaged* and no longer contributes or is removed from the food web.

Most real ecosystems have both points of vulnerability and nonlinear properties, even if the particulars are unknown. However, an additional problem is that many of the mathematical and statistical techniques designed to elucidate cascade behavior have methodological limitations as to the number and type of such vulnerabilities or nonlinear properties they can handle simultaneously. These limitations give rise to production of mathematical artifacts or the predictions or behavior that are not confirmed by observations or measurements on the system being studied.

Because the cascade concept integrates biological and chemical behavior of species communities, it permits simultaneous consideration of the distribution of radionuclides in ecosystems with the effects on ecosystems as modified, if necessary, by the presence and quantities of other biotic and abiotic factors. For the Great Lakes, the cascade concept has been mainly applied to the dynamics of fisheries as influenced by changes in phytoplankton, zooplankton, and selective macroinvertebrate benthos compartments.

PART II

BIOACCUMULATION OF SPECIFIC ELEMENTS

This section addresses individual elements and nuclides with respect to bioaccumulation and biomagnification factors for freshwater biota. The data presentations draw from many studies for the Great Lakes and other locations. A few selected data are presented for terrestrial species of radionuclide importance.

A secondary purpose of this work is to provide an extensive data base of elemental compositions from which to discern appropriate values of bioaccumulation and biomagnification factors radionuclides of various elements and to confirm which factors from tables from *Radioactivity in the Marine Environment* (National Academy of Sciences 1971); the listing of Chapman *et al.* are the most appropriate for Great Lakes work.

TRITIUM

Because of its widespread presence in the effluents (both atmospheric and aquatic) of nuclear power plants, tritium was discussed in detail in the *Inventory* report. Most of the Task Force data were taken from a symposium volume prepared by the IAEA, *Behaviour of Tritium in the Environment*. The symposium volume paper has three valuable papers on tritium uptake by aquatic biota: a paper by Blaylock and Frank (1979) on some plants and animals from a pond on the Oak Ridge Reserve (Tennessee), including two invertebrates and three warmwater fishes; a paper of Kirchmann *et al.* (1979) on several freshwater and marine species to the array, including a salmonid species and important information on the distribution of tritium in cellular compartments; and lastly, a paper by Adams et al. (1979) on tritium uptake by the aquatic biota in a freshwater marsh system in Lake Erie and near the Davis–Besse nuclear power plant. This third paper considers organisms found in the Great Lakes.

These papers showed that tritium levels in organisms track the tritium levels of the environment. Bioaccumulation factors of unity for tritium are conservative. Some investigators note that tritium in organismal tissues quickly "equilibrates" with the tritium content of the surrounding water, but that view is a bit simplistic because "equilibration" masks isotope exchange of tritium from bound water to stable hydrogen on biomolecules. From the available information, a bioaccumulation factor of unity (1) is probably conservative.

GROUP Ia AND Ib ELEMENTS: SODIUM, POTASSIUM, LITHIUM, RUBIDIUM, CESIUM, FRANCIUM AND COPPER, SILVER, AND GOLD

The univalent positive nuclides belong to Groups Ia and Ib of the Periodic Table and consist of sodium, potassium, lithium, rubidium, cesium, and francium (Group Ia) and copper, silver, and gold (Group Ib). Francium is an artificial element, which might be important in high-level waste production, but there is no indication that it is released to the Great Lakes and thus requires no inventory.

Copper and gold bioaccumulate and have important biological properties. However, they do not have long-lived radionuclides that require inventories. The main radionuclide of copper, ^{64}Cu, which occurs as an activation product and from fission product, has a half-life of only 12.5 hours. Thus, the Task Force considers only sodium, potassium, lithium, rubidium, cesium, and silver.

Sodium and potassium

Biologically, potassium and sodium are major elements in tissues (macronutrients). Sodium levels typically exceed potassium levels in natural media (freshwaters, estuaries, and marine waters) as well as in the tissues of vertebrates. The situation typically reverses in aquatic plants and invertebrates with potassium exceeding sodium. This reversal of enrichment acquires special significance given that levels of sodium and potassium in biological tissues are typically of the order of parts per hundred or per cent, rather than parts per million for the trace elements.

Two radionuclides of sodium, ^{22}Na and ^{24}Na, result from both cosmogenic processes and fission, although fission yields are relatively low compared with the levels of cosmogenic production. At least for ^{22}Na, the cosmogenically produced nuclide is believed to contribute to the background dose of radiation received by biota (UNSCEAR 1977). Therefore, some portion of the sodium content of biota may be radioactive. Further, environmental studies of Group Ia elements may require some comparisons with sodium.

The naturally occurring radionuclide of potassium, ^{40}K, is important in any assessment of radioactivity for the Great Lakes. It accounts for as much as 40% of the radioactivity present in all biological tissues (human included), and is one of the two major nuclides that contribute to the natural background dose of radiation, the other one being ^{14}C. Biological measurements of ^{40}K in Great Lakes biota are available from various monitoring studies, and pertinent data appear in several tables that discuss nuclides monitored in the vicinity of discharges from nuclear facilities or in the discussions of other nuclides according to how the original investigators obtained and reported their findings.

Tables 6 and 7 presents Cowgill's data on the uptake of sodium, potassium, lithium, cesium, rubidium, silver, chlorine, bromine, and iodine in aquatic biota. These are the univalent elements of concern in this report. The tables include calculations of bioaccumulation and biomagnification factors. The tabular presentation aims to maintain some integrity of her various data sets as well as present the maximum elemental data economically. Only sodium and potassium are discussed here. The remaining elements are discussed later in their own subsections.

Note that the enrichment of sodium and potassium in the tissues of the two species of plants presented in Tables 6 and 7 shows a reversal of what is observed in source materials. Further note that the plants tend to keep the sodium in the stems and stalks rather than the leaves and flowers. If the aphids (*Rhopalosiphum nymphaea*) consume mainly the leaves and flowers, they are denied a major source of sodium. The theories interpreting the observed sodium enrichment and depletion are not discussed herein because they do not relate to the calculation of inventories for radionuclides of sodium in the Great Lakes.

From Tables 6 and 7, it appears that the bioaccumulation factors for sodium in plant parts are of the order of magnitude of 100–2500 relative to the aquatic environment and the biomagnification factor for the aphid relative to the plant parts is about 1–4. For potassium the bioaccumulation factors for the plant parts relative to the water are 1000–3000 and for the aphid relative to the plant parts is about 1–2.

TABLE 6
UPTAKE OF UNIVALENT ELEMENTS: Na, K, Li, Rb, Cs, Ag, Cl, Br, AND I BY AQUATIC BIOTA
PART I. LINSLEY POND (adapted from the data of Cowgill)

Data for these species **Species and substrates**					**Elements (ppm)**				
	Na	**K**	**Li**	**Rb**	**Cs**	**Ag**	**Cl**	**Br**	**I**
Soils	11,000	11,800	3.5	25.3	1.94	0.02	250.5	5.9	0.14
Rocks	19,700	8,200	6.3	37.6	1.46	0.01	127.3	3.9	0.07
Deep water sediments	10,500	16,600	2.2	47.0	1.35	0.52	478.5	7.7	0.16
Sediments at *Nymphaea odorata*	11,200	210	0.39	27.9	0.52	0.23	434	29.2	0.49
Outlet water	17	9.7	0.0008	0.009	nd	0.0012	5.6	0.014	0.0041
1972 study:									
Nymphaea odorata (water lily)									
Flowers	2600	32,500	0.20	31.1	0.58	0.94	14,230	30.6	0.52
Flower stalks	31,700	28,600	0.15	23.1	0.48	0.24	28,790	41.3	0.52
Leaves	3200	22,400	0.30	21.9	0.67	0.26	13,560	26.2	0.54
Stems	28,200	25,900	0.15	23.7	0.46	0.20	31,920	44.8	0.56
1971 study:									
Nymphaea odorata (water lily)									
Flowers	3600	24,300	0.27	29.0	0.64	0.66	14,680	65.6	0.37
Flower stalks	30,000	18,400	0.23	25.1	0.54	0.30	27,560	54.1	0.38
Leaves	2100	14,200	0.27	24.2	0.75	0.32	14,940	38.2	0.58
Stems	36,300	19,600	0.16	24.7	0.67	0.30	40,580	62.6	0.55
Rhopalosiphum nymphaeae (aphid)	7100	22,400	1.6	28.4	1.23	0.42	2090	17.5	0.66

TABLE 6 (continued)
UPTAKE OF UNIVALENT ELEMENTS: Na, K, Li, Rb, Cs, Ag, Cl, Br, AND I BY AQUATIC BIOTA
PART I. LINSLEY POND (adapted from the data of Cowgill)

Bioaccumulation and biomagnification factors

	Na	K	Li	Rb	Cs	Ag	Cl	Br	I
Plants/water: 1971 study (R_1)									
Flowers	212	2505	3380	3222	*	53,883			
Flower stalks	1765	1897	2875	2789	*	25,000			
Leaves	123	1464	3380	2689	*	26,667			
Stems	2135	2021	2000	2744	*	25,000			
Aphids/plants: 1971 study (R_3)									
Flowers	1.97	0.92	5.9	0.97	1.92	0.64			
Leaves	3.38	1.58	5.9	1.02	1.64	1.32			

Geochemical ratio calculations

	Na/K	Li/K ($\times 10^4$)	Rb/K ($\times 10^{-3}$)	Cs/K ($\times 10^{-4}$)	Br/Cl ($\times 10^{-3}$)	I/Cl ($\times 10^{-5}$)
Soils	0.932	2.96	2.16	1.64	23.55	55.9
Rocks	2.42	7.68	4.58	1.78	30.64	55.0
Deep water sediments	0.63	1.32	2.83	0.813	16.09	33.4
Sediments at *Nymphaea odorata*	53.3	18.5	13.2	2.47	67.28	112.9
Outlet water	1.75	0.82	9.28	*	2.50	73.2
Nymphaea odorata (average of 1971–1972 data)						
Flowers	0.109	0.083	15.8	2.53	3.33	3.11
Flower stalks	1.31	0.081	11.0	2.19	1.69	1.59
Leaves	0.145	0.156	14.2	3.63	2.26	3.92
Stems	1.42	0.068	11.6	2.65	1.48	1.54
Rhopalosiphum nymphaeae	0.32	0.714	12.6	0.055	8.37	31.57

Notes: (1) Substrate data are averages of the results of years 1971–1972. Plants were collected in both 1971 and 1972; aphids *(Rhopalosiphum nymphaea)* were collected only in 1971.

(2) Calculation of bioaccumulation and biomagnification ratios based on the year indicated for the plant data and the averaged data for water or source materials.

(4) Symbols and abbreviations: "nd" means not detected. The asterisk (*) means no computation for the quantity in the table. In this case the quantity, the bioaccumulation factor for cesium, cannot be computed mathematically. Since cesium was not detected in the outlet water of the pond, that means that the cesium content of the water could be "zero." That would produce an estimate of an "infinite" bioaccumulation factor, a mathematical absurdity.

(4) It is not geochemically appropriate to compare silver with potassium because silver is in Group 1b of the Periodic Table. Cowgill compared silver to copper in the same group, but the copper data have not been presented here. Thus silver is included in the geochemical ratio calculations of the univalent positive elements of Group 1.

TABLE 7
UPTAKE OF UNIVALENT ELEMENTS: Na, K, Li, Rb, Cs, Ag, Cl, Br, AND I BY AQUATIC BIOTA
PART II. CULTURES (adapted from the data of Cowgill)

Elements (ppm)

Species or substrate	**Na**	**K**	**Li**	**Rb**	**Cs**	**Ag**	**Cl**	**Br**	**I**
Euglena gracilis	252	17,898	0.60	26.4	1.77	4.7	360	18.0	1.75
Mixed algal culture	200	13,955	0.42	20.7	1.12	4.6	400	14.2	1.40
Daphnia pulex	861	8003	0.38	35.3	0.82	0.78	3198	27.1	0.54
Daphnia magna	1180	13,839	0.28	22.9	0.28	0.46	10,130	37.1	0.69
Average *Daphnia*	1020	10,921	0.33	29.1	0.55	0.62	6664	32.1	0.62
Average algae	226	15,927	0.51	23.6	1.45	4.6	380	16.1	1.58
Trap rock	15,753	5562	6.23	18.9	1.26	0.009	128	3.8	0.035
Spring water ($\times 10^4$)	1130	2580	2.0	19.0	0.56	0.068	1450	200	6.7

Bioaccumulation and biomagnification factors

Species or substrate	**Na**	**K**	**Li**	**Rb**	**Cs**	**Ag**	**Cl**	**Br**	**I**
Euglena gracilis / spring water	2230	69,372	3000	13,894	31,607	691,176	2482	900	2612
Mixed algal culture / spring water	1769	54,089	2100	10,895	20,000	676,470	2759	710	2090
Daphnia pulex / spring water	7619	31,020	1900	18,579	14,642	114,705	22,055	1355	806
Daphnia magna / spring water	10,442	53,640	1400	12,052	5000	67,647	69,862	1855	1029
Average *Daphnia* /spring water	9027	42,329	1650	1532	9821	91,176	45,959	1605	918
Daphnia pulex / mixed algal culture	4.3	0.6	0.9	1.71	0.73	0.17	7.99	1.91	0.39
Daphnia magna / mixed algal culture	5.1	0.99	0.67	1.11	0.25	0.10	25.3	2.61	0.49
Average *Daphnia* / mixed algal culture	4.7	0.79	0.79	1.41	0.54	0.14	16.6	2.26	0.44

TABLE 7 (continued)
UPTAKE OF UNIVALENT ELEMENTS: Na, K, Li, Rb, Cs, Ag, Cl, Br, AND I BY AQUATIC BIOTA
PART III. OTHER STUDIES (adapted from the data of various authors)

Elements (ppm)

Species or substrate	Na	K	Li	Rb	Cs	Ag	Cl	Br	I
Victoria amazonica									
Young plants	2074	10,631	1.14	0.047	15.6	0.059	1946	6.1	0.036
Pre-flowering plants	3165	21,461	0.662	0.049	22.3	0.065	6595	10.5	0.273
Mature plants; unopened buds	3936	26,667	0.639	0.051	27.8	0.074	8667	15.0	0.293
Mature plants; full bloom	4972	29,460	0.551	0.063	32.4	0.074	9873	17.8	0.345

Geochemical ratios

	Na/K	Li/K ($\times 10^5$)	Cs/K ($\times 10^6$)	Rb/K ($\times 10^3$)	Br/Cl ($\times 10^4$)	I/Cl ($\times 10^5$)
Young plants	0.195	10.7	4.5	1.47	8.85	1.85
Pre-flowering plants	0.147	3.08	2.3	1.03	15.9	4.14
Mature plants; unopened buds	0.148	2.4	1.9	1.04	17.3	3.38
Mature plants; full bloom	0.169	0.187	2.1	11.0	18.0	3.49

Notes: (1) Reference: Cowgill and Prance (1982)
(2) All data are based on dry weight
(3) Geochemical ratios are not provided for silver because it is not appropriate to calculate them with potassium as a reference element.

Lithium

Lithium has small enough atomic and ionic radii to replace sodium and potassium in various chemical matrices. The element has a metabolic role with respect to neuron function in animal cells. Because it can produce radionuclides of beryllium under a number of different simple nuclear reactions and is itself sometimes the stable daughter product of the radioactive decay of certain beryllium radionuclides, it would appear logical to treat lithium and beryllium together. However, their geochemical coherence is not obvious in biological materials, and therefore, lithium is discussed with the other univalent positive elements, and beryllium, with divalent positive elements.

Lithium data for plants are limited. Cannon's work (1960) on lithium accumulator plants for geobotanical prospecting is important for terrestrial species. Cowgill's work is probably the most important and complete data set for lithium in aquatic species. Her lithium data are included in Tables 6 and 7.

Rubidium

Rubidium has sufficiently small atomic and ionic radii that it can substitute for potassium and sodium in some chemical matrices. Rubidium's geological rarity somewhat limits its biological availability.

The substitution of rubidium for potassium or sodium suggests a possible environmental risk factor associated with its uptake and the likelihood that it will bioaccumulate to greater levels than cesium. Existing evidence supports both statements. Rubidium accumulates to levels about 100 times greater than cesium. Rubidium has acted as a growth stimulant and electrolyte balancing element in tissues that were temporarily depleted or impoverished with potassium (Luckey and Venugopal 1978).

There are many important radionuclides of rubidium: ^{83}Rb, ^{85}Rb, ^{86}Rb, ^{87}Rb, and ^{88}Rb. The first three nuclides come either directly from fission of uranium or as an immediate daughter isotope of another direct fission product. Thus, they appear in the effluents of nuclear power plants either directly or indirectly. The fourth radionuclide is primordial. Assessments of the health risk from rubidium uptake usually examine the primordial nuclide because it is assumed to form a significant part of the natural background radioactivity to which all life is exposed. Yet according to information provided by UNSCEAR (1977) reports, the nuclide was not itself the subject of any uptake or metabolic studies. Rather, data from studies with stable rubidium isotopes formed a basis to infer the metabolic risks associated with the naturally occurring radionuclide.

Radionuclides of rubidium are not measured in biota and bioconcentration factors for them do not appear in the two main lists previously cited in this chapter. Therefore, one must rely on data from other studies, of which Cowgill's data are the most comprehensive early data.

Cesium

Although its chemistry parallels that of sodium and potassium, cesium does not routinely replace either sodium or potassium in various chemical matrices. Cesium's atomic and ionic radii are considerably larger than those of sodium and potassium, sometimes making such replacement difficult. This large size of the cesium nucleus would thus suggest a rather small biotic uptake for the element. Indeed, the data from Cowgill given Tables 6 and 7 suggest 0.1–5 parts per million (ppm) levels, but the rarity of cesium as an element in the earth's crust also limits its biological availability and could imply that the observed ppm levels are higher than might otherwise be expected.

Cesium's radionuclides are important for two reasons:

(1) They have atomic weights in a range highly favored in the kinetics of nuclear fission of uranium and plutonium. Therefore, cesium isotopes occur in significant quantities in the nuclear debris from weapons testing and in the liquid effluents of nuclear reactors.

(2) They are decay products of noble gas radionuclides of xenon, which are produced in fission processes. Again, cesium isotopes will be present in the nuclear debris from weapons testing and formed from decay of the isotopes in the gaseous emissions of nuclear power plants.

Very few radionuclide tracer studies on freshwater species have emphasized long-lived artificial nuclides. Early radionuclide tracer studies used ^{32}P (half-life 14.28 days) because this nuclide was the first to become commercially available to research scientists. Other studies used the long-lived ^{14}C (half-life 5730 years), especially in work on the "geochronology" (dating) of the materials, and some work on the environmental cycling of carbon. ^{137}Cs (half-life 30 years) became the first long-lived isotope related to nuclear fallout intensively studied in aquatic organisms. Given the 30-year half-life of the isotope, it became a prime candidate for biological monitoring in studying radiation effects.

^{137}Cs emits a distinctive γ radiation signal that permits its analysis by gamma radiation spectroscopy. The nuclide is usually detectable if present, and it is often the only artificial nuclide which is detected in certain types of samples. Thus, the quantity of data for this nuclide probably exceeds that of all nuclides monitored from fallout.

The early studies of cesium uptake emphasized marine biota as part of the tracking of the element to ocean repositories. Very few definitive studies exist for cesium uptake in freshwater biota. Some studies of cesium uptake in algae in special cultures came from researchers at the nuclear weapons laboratories, but studies of radiocesium uptake in macrophytes and aquatic invertebrates are far fewer. The most famous work on radiocesium accumulation is the study of the uptake of cesium from nuclear fallout in the Arctic by lichens, the consumption of the lichens by reindeer or caribou, and the exposure of human populations who consume the cesium-contaminated meat.

Given the previous information on the extent of studies using ^{137}Cs, it should not be surprising that very high-quality bioaccumulation data for this nuclide are available in research studies. In fact, the Task Force considers the research data for bioaccumulation of this nuclide to be the most reliable of all of the bioaccumulation data for the artificial radionuclides.

Geochemical interpretations of cesium data often use its congener element, potassium, as a reference element. This permits the consideration of the possibility that cesium and potassium exhibit "coherence," a geochemical term which suggests that two elements move together environmentally or follow similar kinetic processes and mechanisms even if they do not move together environmentally. Since potassium has a major role in plant and animal metabolism, the comparisons may provide insight into the metabolic behavior of cesium relative to potassium. Cowgill found that relative to potassium, cesium accumulation in aquatic plants exhibited a geochemical ratio (Cs/K) of 0.25×10^{-4}. Taylor (1964) suggested that the cesium to potassium geochemical ratio of the "accessible lithosphere" was 1.4×10^{-4}. Thus, while cesium clearly accumulates in biological materials, it does not necessarily accumulate to levels in excess of that found naturally in the source mineral substrates. Also, while the two geochemical ratios have the same order of magnitude, they differ sufficiently to suggest that cesium uptake does not occur mechanistically by processes that mimic uptake and behavior of potassium. The differences are in the metabolism of the two elements. Since cesium does not necessarily substitute for potassium, cesium's metabolic behavior is more limited than that of potassium.

Studies on radioactive cesium in plants have included environmental behavior in a variety of species: ranging from cesium uptake by algae in laboratory cultures, to the cesium levels in "periphyton" (reported by some nuclear power plant dischargers in their monitoring work) to extensive research data on terrestrial plants (*i.e.*, forest stands, agricultural plots) and emergent wetland plants (those that root in a mud or aquatic system but emerge and grow in the terrestrial environment or at the water's edge). The most comprehensive of the early studies of the 1970s were reported in various conference proceedings of the United States Department of Energy.

The deposition of radionuclides of cesium from the atmosphere in the northern hemisphere favors the northernmost latitudes. Some of the ecological systems in these latitudes are rather limited because of the climate conditions. An important system in these regions is the lichen – reindeer/caribou – human food chain. Lichens, a plant–fungus symbiont, can readily accumulate metallic elements because some of the plant material behaves like an ion-exchange resin. This food chain pathway to man of the radionuclides of cesium has been studied in various Arctic regions, including northern Canada, Alaska, Denmark, the Faroë Islands, Sweden, Norway, Finland, and Commonwealth of Independent States (formerly Russia). Some of the data on caribou are presented in Table 8. Following the accident at the nuclear power plant at Chernobyl (Ukraine) in 1986, researchers and Government agencies undertook new studies of atmospheric deposition in the Arctic. Many of the results of these studies appeared in 1995 in a special symposium on pollution in the Arctic (Volume 160/161, *Science of the Total Environment*). The research findings updated important information on ^{137}Cs levels in Arctic species, including some species not previously considered.

TABLE 8
ELEMENTAL ANALYSES (INCLUDING RADIONUCLIDES) OF ARCTIC BIOTA

Study: Elkin and Bethke (1995)
Contaminants in caribou from Northwest Territories, Canada

Species: caribou (*Rangifer tarandus*) muscle tissue

Station	Nuclide (Bq/kg)		Station	Nuclide (Bq/kg)		Station	Nuclide (Bq/kg)	
Bathurst	^{137}Cs	46.98	Cape Dorset	^{137}Cs	51.24	Lake Harbour	^{137}Cs	185.48
	^{134}Cs	<0.49		^{134}Cs	0.64		^{134}Cs	<2.08
	^{40}K	155.2		^{40}K	153.7		^{40}K	171.23

Geochemical ratios

^{137}Cs/^{40}K	Bathurst	0.303	Cape Dorset	0.333	Lake Harbour	1.08	Average of all sites	0.572
^{134}Cs/^{40}K				0.004				
(^{134}Cs + ^{137}Cs)/^{40}K				0.338				

Notes: (1) The 13 lichens are those for which there are both Sr and Cs data. The 14 lichens are for the entire collection of lichens. The geochemical ratios are based on 13 lichens and 15 mosses. The combined geochemical ratio for lichens and mosses is based on 28 plant species.

(2) Nuclide data are on a dry weight basis. However, Elkin and Bethke indicate that the water contents of the muscle samples were 25.8% (Bathurst), 24.7% (Cape Dorset), and 27.7% (Lake Harbour) and gave their data in wet weight. The nuclides have been recalculated from this information.

(3) Results are means of 20 animals sampled in the Bathurst herd and 10 animals sampled from each of the Cape Dorset and Lake Harbour herds.

(4) The Lake Harbour herd is also the highest of the sampled herds for aluminum, cadmium, chromium, manganese, nickel, lead, and mercury in both kidney, and liver tissue. These results are given in other tables in this report.

As previously noted, there is no inherent reason to assume that either Sr and Cs will bioaccumulate in plants in the same ratios as they appear in nuclear fallout. In fact, in aquatic species of interest to the Great Lakes, the accumulations of these two elements differ by at least an order of magnitude. Thus, it may seem surprising that the two major radionuclides of Sr and Cs from fallout appear to accumulate in mosses and lichens in ratios that track their relative distribution in nuclear fallout. Because mosses and lichens behave like living versions of ion-exchange resins and can exchange strontium and cesium ions for hydrogen ions. A two-step accumulation process occurs: a stoichiometric binding as a rapid and dominant first step, and possible incorporation of a nuclide into tissue as a second step. The Task Force would not expect this ion-exchange behavior in Great Lakes biota.

The important compilations on radiocesium uptake come from the work of Blaylock (1982), Joshi (1984), and Hesslein and Slaviček (1984). The latter two references uniquely address Great Lakes fishes. Some of the materials in those compilations come from monitoring studies, but much of the data collection was research motivated.

Tables 9 and 10 present data from previous IJC compilations on radioactivity in the Great Lakes with respect to bioaccumulation studies for ^{137}Cs and includes some limited citations from Joshi's (1984) paper for the bioaccumulation factors. The original data compilations presented both mass of the fish collected and their activity per unit mass. The activity per unit mass stays relatively constant over a fourfold range of mass (1.5–6.0 kg) of the fish. Further, the mean activity per unit mass for fishes in the two lakes are within a similar range of numbers, although the mean activities for the two species shown for Lake Huron are not identical by a two-tailed *t* test, while the mean activities for the species in Lake Ontario are likely the same by the two-tailed *t* test. Text Box 6 shows the dosages in microsieverts that would be ingested by a person eating the affected fish.

The Ganaraska River flows into Lake Ontario and carries the effluents from the CAMECO fuel processing site. Since the cesium activity data from the fish in the river do not differ strongly from the cesium activity of the data from fishes within Lake Ontario stations, the Task Force suggests that the source of radiocesium is probably nuclear fallout and not direct radionuclide discharge from the CAMECO operations. The radium data for Ganaraska River and the other Lake Ontario stations (not presented here) do show specific source inputs of the CAMECO site.

TABLE 9
CESIUM ACCUMULATION IN GREAT LAKES BIOTA
PART I — ^{137}Cs IN GREAT LAKES FISHES
(adapted from International Joint Commission (1983, 1987), Joshi (1984), and other agency reports)

Year	Species	Location	Average ^{137}Cs Activity (pCi/kg) factor	Bioaccumulation
Lake Huron:				
1981	Walleye	Blind River	264.5 ± 11.3 (6)	
1981	Sturgeon	Blind River	90.8 ± 6.8 (4)	
1982	Lake trout	North Channel	222.3 ± 4.7 (3)	
Lake Erie:				
1982	Walleye	Western Basin	23.3 ± 2.3 (3)	1556
Lake Ontario:	**Ganaraska River**			
1976	Rainbow trout		64	3528
1977	Rainbow trout		53	2391
1978	Rainbow trout		60	2354
1980	Rainbow trout		72	
1981	Rainbow trout		37.8 ± 3.6 (9)	1700
Lake Ontario:	**Other Locations**			
1982	Rainbow trout	Coburg	41 ± 3	1414
1982	Lake trout	Coburg	41.3 ± 3 (3)	1425
1982	Lake trout	Niagara on the Lake	43.3 ± 3.3 (4)	1490
1982	Lake trout	Oswego	45 ± 10 (2)	

Notes: Numbers in parentheses indicate number of fishes in sample used in the averaging. If no number appears in parentheses, then either only one measurement was reported, or the source of the data did not qualify the information in some manner. This is especially important for the data on the Ganaraska River for 1976 to 1978, where the sources used reported means but not uncertainties or standard errors in the compilation. Radioactive measurements were made on a wet weight basis. Data are from both Canadian (IJC 1983, 1987; Joshi 1984; Environment Canada, personal communication) and United States (New York State Department of Health 1983–1993) sources. Bioaccumulation factors from Joshi (1984) relate to open water levels or ambient levels at collection site, depending on the available data.

All of the bioaccumulation factors cited have the same order of magnitude, although fishes in Lake Huron have higher bioaccumulation factors than fishes in Lake Ontario. These factors should not be assumed to hold for the two Lakes over all periods of time. Joshi (1984) calculated bioaccumulation factors from several fish studies going back to 1976 (some of the data from which appear in Table 10). His calculations suggest that bioaccumulation factors were much higher in early years and have exhibited a downward trend with time. His work also indicated that fishes in lakes not receiving direct discharges of radionuclides (Lake Superior) sometimes had higher bioaccumulation factors than fishes from lakes that had direct discharges of radionuclides. There are several possible explanations for such an observation, but because the data bases are not sufficiently extensive to perform a statistically comprehensive interlake comparison of bioaccumulation factors of cesium or test any hypotheses or reasons for trends or differences in apparent factors from lake to lake, the Task Force cautions that no particular emphasis should be placed on any trend suggested by the data; they merely illustrate what has been reported and observed.

TABLE 10
CESIUM ACCUMULATION IN GREAT LAKES BIOTA
PART II — ^{137}Cs IN GREAT LAKES FISHES: STUDIES AT HWR NUCLEAR GENERATING STATIONS

(adapted from operating reports of Ontario Hydro)

Year	Lake	HWR and Station	Sample or Substrate	Activity (Bq/L or Bq/kg)		^{137}Cs/^{40}K
				^{137}Cs	^{40}K	
1992	Ontario	Darlington (Provincial Park)	Trout	1.07 ± 0.15	120 ± 4	0.0089
			Whitefish	0.56 ± 0.15	119 ± 4	0.0047
			Sucker	0.18 ± 0.15	105 ± 6	0.0017
			Water	<0.003	0.14 ± 0.03	
			Sediments	2.5 ± 1.3	360 ± 30	0.0069
		Bioaccumulation factors:	Trout/water	*	857	
			Whitefish/water	*	850	
			Sucker/water	*	750	
1992	Ontario	Darlington (NGS)	Trout	0.96 ± 0.18	124 ± 6	0.0077
			Whitefish	0.48 ± 0.18	126 ± 6	0.0038
			Sucker	0.23 ± 0.11	109 ± 4	0.0021
			Water	<0.003	0.14 ± 0.04	
			Sediments	<1.2	410 ± 30	
		Bioaccumulation factors:	Trout/water	*	886	
			Whitefish/water	*	900	
			Sucker/water	*	779	
1992	Ontario	Pickering ("A" discharge)	Trout	1.04 ± 0.22	137 ± 7	0.0076
			Whitefish	0.63 ± 0.15	120 ± 4	0.0053
			Sucker	1.15 ± 0.22	121 ± 7	0.0095
			Water	0.015 ± 0.004	0.14 ± 0.04	0.107
			Sediments	16.5 ± 1.5	410 ± 30	0.0402
		Bioaccumulation factors:	Trout/water	69.3	979	
			Whitefish/water	42.0	857	
			Sucker/water	76.7	864	

TABLE 10 (continued)
CESIUM ACCUMULATION IN GREAT LAKES BIOTA
PART II — ^{137}Cs IN GREAT LAKES FISHES: STUDIES AT HWR NUCLEAR GENERATING STATIONS
(adapted from operating reports of Ontario Hydro)

Year	Lake	HWR and Station	Sample or Substrate	Activity (Bq/L or Bq/kg) ^{137}Cs	^{40}K	^{137}Cs/^{40}K
1992	Ontario	Pickering ("B" discharge)	Trout	0.92 ± 0.18	132 ± 4	0.0069
			Whitefish	0.85 ± 0.22	136 ± 6	0.0063
			Sucker	1.00 ± 0.22	131 ± 6	0.0076
			Water	<0.003	0.17 ± 0.05	
			Sediments	7.0 ± 1.3	430 ± 30	0.0162
		Bioaccumulation factors:	Trout/water	*	776	
			Whitefish/water	*	800	
			Sucker/water	*	771	
1992	Ontario	Pickering (Duffin's Creek)	Trout	1.00 ± 0.15	104 ± 3	0.0096
			Whitefish	0.74 ± 0.22	133 ± 6	0.0056
			Sucker	0.96 ± 0.18	153 ± 4	0.0063
			Water	<0.003	0.17 ± 0.04	
			Sediments	8.5 ± 1.4	390 ± 20	0.022
		Bioaccumulation factors:	Trout/water	*	612	
			Whitefish/water	*	783	
			Sucker/water	*	900	
1992	Ontario	Pickering (Coldwater Farms)	Rainbow trout	<0.15	128 ± 4 (w)	
			Rainbow trout	0.2 ± 0.1	117 ± 4 (s)	0.0017

TABLE 10 (continued)
CESIUM ACCUMULATION IN GREAT LAKES BIOTA
PART II — ^{137}Cs IN GREAT LAKES FISHES: STUDIES AT HWR NUCLEAR GENERATING STATIONS
(adapted from operating reports of Ontario Hydro)

Average geochemical ratios ^{137}Cs/^{40}K factors for species monitored at HWR facilities

HWR	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K	Species	^{137}Cs/^{40}K
Lake Ontario:								
Darlington	Trout	0.0083	Whitefish	0.0043	Sucker	0.0019		
Pickering	Trout	0.0083	Whitefish	0.0053	Sucker	0.0078		
Average for Lake Ontario fish:								
	Trout	0.0083	Whitefish	0.0048	Sucker	0.0048	All fishes	0.0060

Average bioaccumulation factors for species monitored at HWR facilities

		^{137}Cs	^{40}K	^{137}Cs/^{40}K	Lake Ontario:		^{137}Cs	^{40}K
Lake Ontario:					Pickering			
Darlington								
	Trout/water	*	872			Trout/water	69	789
	Whitefish/water	*	875			Whitefish/water	42	813
	Sucker/water	*	765			Sucker/water	77	845
	Average all fish/water	*	837			Average all fish/water	63	816
Lake Huron: 1992								
Bruce (Discharge "A")								
	Walleye	2.78 ± 0.27	105 ± 5	0.0264				
	Bass	3.03 ± 0.19	121 ± 4	0.025				
	Sucker	1.48 ± 0.15	138 ± 4	0.010				
	Water	<0.003	0.11 ± 0.03					
Bioaccumulation factors:	Walleye/water	*	954					
	Bass/water	*	1100					
	Sucker/water	*	1254					
Bruce (Discharge "B")								
	Walleye	2.74 ± 0.19	129 ± 4	0.0212				
	Bass	2.15 ± 0.30	126 ± 6	0.017				
	Trout	1.52 ± 0.19	152 ± 4	0.01				
	Water	<0.003	0.11 ± 0.04					
Bioaccumulation factors:	Walleye/water	*	1172					
	Bass/water	*	1145					
	Trout/water	*	1381					
Bruce (Discharge "A")								
	Trout	3.33 ± 0.19	123 ± 4	0.027				
	Pike	1.96 ± 0.15	105 ± 3	0.0187				
	Carp	1.33 ± 0.15	88 ± 3	0.015				

TABLE 10 (continued)
CESIUM ACCUMULATION IN GREAT LAKES BIOTA
PART II — ^{137}Cs IN GREAT LAKES FISHES: STUDIES AT HWR NUCLEAR GENERATING STATIONS
(adapted from operating reports of Ontario Hydro)

Average geochemical ratios for $^{137}Cs/^{40}K$ for species monitored at HWR facilities

HWR	Species	$^{137}Cs/^{40}K$	Species	$^{137}Cs/^{40}K$	Species	$^{137}Cs/^{40}K$	Species	$^{137}Cs/^{40}K$
Lake Huron:								
Bruce	Trout	0.0185	Walleye	0.0238	Bass	0.021	All fish	0.0194

Average bioaccumulation factors for species monitored at HWR facilities

	^{137}Cs	^{40}K
Lake Huron:		
Bruce		
Walleye/water	*	1063
Bass/water	*	1123
Trout/water	*	1318
Average all fish/water	*	1168

Comparisons of Lakes Ontario and Huron

Average geochemical ratios for $^{137}Cs/^{40}K$

Lake Ontario/Lake Huron	
Trout	0.45
All fish	0.31

Average bioaccumulation factors ^{137}Cs and ^{40}K

	^{137}Cs	^{40}K
All fish/water	*	1.41

Notes: (1) Sediment data are dry weight; fish data are wet weight

(2) Symbols: (w) samples taken in winter–spring period (January to June); (s) samples taken in summer–autumn period (July to December); (*) cannot calculate the number from information given.

(3) Averages of bioaccumulation factors are calculated when there are two or more results for a given species of fish. The overall average bioaccumulation factor for fishes in a lake averages all data from that lake without regard to the number of entries per species.

(4) Bioaccumulation factors reported for ^{137}Cs based on one data set only

(5) Ratio of bioaccumulation factors for ^{40}K for Lake Ontario and Lake Huron comparison obtained by taking the ratio of the averages of all of the bioaccumulation factors for the fishes of each lake.

TEXT BOX 6
RADIATION DOSES RESULTING FROM RADIONUCLIDE LEVELS IN FISH (TABLE 10)

Species of fish	Range of ^{137}Cs concentrations (Bq/kg)	Range of doses (μSv/year)
Trout	0.92–3.33	1.2–4.3
Whitefish	0.48–0.85	0.6–1.1
Sucker	0.18–1.48	0.2–1.9
Walleye	2.74–2.78	3.6
Bass	2.15–3.03	2.8–3.9
Carp	1.33	1.7

Notes: (1) Dose estimates are for ^{137}Cs only. ^{40}K levels in the body are homeostatically controlled, *i.e.*, if more comes from one source, less is accepted from other sources.
(2) Doses are calculated based on the assumption that an adult consumes 100 kg/year of fish (about 0.5 pounds/day). The conversion factor is 0.013 microsieverts (μSv) per becquerel (Bq) ingested.

A simple comparison of the data from Tables 9 and 10 shows that Arctic species accumulate radionuclides of cesium over two orders of magnitude greater than Great Lakes species. Unfortunately, data for Lake Superior are not present. This is the largest of the Great Lakes with the largest surface area. Since fallout increases with surface area, it is difficult to tell whether Arctic species have a greater radiocesium inventory than Great Lakes species. Arctic species only receive nuclear fallout, while the Great Lakes species receive fallout, gaseous and liquid discharges of nuclear power plants, and the runoff of deposited materials from nearby areas in the watershed. Fallout fluxes depend on latitude band, and the latitude band for the Great Lakes has a higher flux than the latitude band for the Arctic, but the Arctic region has a greater surface for deposition than the Great Lakes. What is clear is that both regions, Arctic and Great Lakes receive a high quantity of radiocesium, and that the biota in both regions reflect this.

Silver

One radionuclide of silver, ^{110m}Ag, consistently appears in the nuclear discharges to the Great Lakes. The nuclide has been reported every year in the gaseous and liquid discharges of at least eight nuclear power plants in the Great Lakes over the period of 1980–1993. Its half-life of 253 days assures that silver can cycle through Great Lakes biota, if bioaccumulated, over a measurable period of a year following its discharge. The isotope decays by β emission to form a stable nuclide of cadmium, but silver and cadmium do not usually move geochemically together; thus, silver is not discussed with cadmium. The isotope also undergoes internal nuclear rearrangement (note the letter "m" designation in the isotope), to produce the isotope ^{110}Ag (half-life 24.4 seconds), which also decays by β emission.

Very little is known about the environmental cycling of silver. It is highly toxic to aquatic microorganisms and fishes. The Task Force decided to include a presentation of data for silver in its report and estimate a biological inventory for its important radionuclide. Again, Cowgill's data are the most comprehensive for freshwaters systems and appeared in Table 7.

GROUP IIa ELEMENTS: BERYLLIUM, MAGNESIUM, CALCIUM, STRONTIUM, AND BARIUM

The Group IIa positive divalent elements are beryllium, magnesium, calcium, strontium, barium, and radium. Radium, a naturally occurring radioactive element is treated separately with the transuranic elements. Data on the uptake of Group IIa elements are given in Table 11.

Beryllium

Beryllium accumulates in plants. Two main radionuclides of beryllium, ^{7}Be and ^{10}Be, were previously discussed because they are cosmogenically produced. When these nuclides originate from cosmogenic processes, they usually do not last long enough to reach freshwater systems and be accumulated by biota, although this can happen under favorable conditions of atmospheric deposition. The nuclides also are produced in nuclear reactors and is a likely source of radioactive beryllium, which can accumulate in plants and animals in the Great Lakes, and possibly from radiation interactions with source materials of the Basin. Indeed, the Task Force has noted the occurrence of ^{7}Be in the effluents (gaseous and liquid) of nuclear power plants. However, animals reject beryllium in the food supply. The calculated bioaccumulation factors for beryllium from the data sets available in this report are of the order of 0.06 for animals relative to plants. Thus, other than a source of internal radiation, the radioactive beryllium levels in tissues would not appear to be a bioaccumulation problem. On the other hand, humans who breathe in particulate matter containing beryllium are at risk from beryllosis and lung cancers. Airborne levels of beryllium are of concern in cancer risk assessments for hazardous air pollutants, but none of the pathways of beryllium exposure have thus far been shown to be significantly impacted by bioaccumulation related processes.

Calcium and Magnesium

Calcium and magnesium are macronutrient elements in biological tissues. They have roles in maintenance of ionic balance, energy metabolism, production of DNA and RNA, and neurological function (animal cells). Calcium is the primary cation in skeletal tissue of vertebrates and invertebrates, and magnesium is the essential inorganic element in chlorophyll.

Neither the radionuclides of calcium nor magnesium require their inventories in biological compartments of the Great Lakes because these nuclides do not have sufficiently long half-lives for consideration. Both elements, however, are needed in discussing the behavior of the radionuclides of other elements cycling within the Great Lakes, especially in developing inventories for the radionuclides of beryllium, strontium, and barium.

Strontium

Strontium can replace calcium in the bone tissue of vertebrates. The radionuclides of strontium are mostly β and γ emitters, making them a major source of internal radiation when incorporated into tissues. In humans and other vertebrates, radionuclides of strontium affect the mineral integrity of bone tissue, and the internal radiation affects bone cells and bone marrow (the blood forming organ found in bones).

Of the several radionuclides of strontium formed in fission either directly or as the decay products of other nuclides (mainly noble gas radionuclides of krypton), ^{90}Sr is especially important because of its long half-life of 30 years. A second nuclide of concern, ^{89}Sr, also a fission product, occasionally appears in nuclear fallout.

Nuclear power plants produce many strontium nuclides. Nuclear power plants in the United States within the Great Lakes Basin reported ^{89}Sr in liquid effluents 117 times during the period of 1980–1993, with at least seven nuclear power plants reporting the isotope each year. The maximum number of United States nuclear power plants in the Great Lakes Basin which reported this isotope in liquid effluents was 11, and that occurred in 1991. All United States and Canadian nuclear power plants report discharges of ^{90}Sr. One or two power plants yearly report releases of ^{92}Sr, and very rarely, perhaps once every 5 or 6 years, a power plant reports releases of ^{91}Sr. Among the other strontium radionuclides reported as fission products for Canadian plants with respect to inventories for high-level waste disposal are ^{86}Sr and ^{87}Sr. The stable nuclide, ^{88}Sr, is reported in high-level waste but not reported in radioactive effluents.

Barium

Only ^{140}Ba is important in inventories of radionuclides. This fission product decays to ^{140}La, and these two nuclides are sometimes treated together. Some sources even report the data for the two isotopes as a combined activity, without indicating which fraction of the activity belongs to each isotope. Only barium is discussed in this section. Lanthanum, a rare earth element, is treated with other rare earth elements, notably radionuclides of cerium.

Barium accumulates only slight in organisms, the most definitive freshwater studies being those of Cowgill. Its accumulation strongly depends on levels of calcium both within the environment and within organism tissues. High calcium levels in tissues tends to block accumulation and deposition of barium.

TABLE 11
UPTAKE OF GROUP IIA AND GROUP IIIA ELEMENTS: Be, Mg, Ca, Sr, Ba, B, AND Al VARIOUS STUDIES (adapted from work of various authors)

Species	**Elements (ppm)**							**Geochemical ratios (×10^4)**			
	Ca	**Mg**	**Be**	**Sr**	**Ba**	**B**	**Al**	**Be/Mg**	**Ba/Ca**	**Sr/Ca**	**B/Al**
Study: Cowgill (1976)											
Euglena gracilis	7311	5996	0.088	40.0	48.5	12.0	190.5	0.147	66.3	54.7	632
Mixed algal culture	9112	6520	0.084	35.3	44.5	17.8	232.9	0.129	48.8	38.7	764
Daphnia magna	76,643	633	0.028	45.8	84.3	14.0	121.7	0.004	11.0	5.98	1157
Daphnia pulex	36,878	549	0.017	81.9	67.1	14.0	127.0	0.005	18.2	22.2	1102
Average algae	8212	6258	0.86	37.6	46.5	14.9	211.7	0.138	57.6	46.7	698
Average *Daphnia*	56,761	591	0.023	63.9	75.7	14.0	124.4	0.0045	14.6	14.1	1130
Spring Water (×10^4)	44,900	18,800	0.15	330	140	580	2070	0.816	31.2	73.5	2801
Trap rock	57,151	40,058	0.123	129.3	126	130	53,179	0.031	22.0	22.6	24.4
	Bioaccumulation factors										
Average algae / spring water	1828	3329	5733	1139	3321	257	1019	1.72	1.82	0.62	0.252
Average *Daphnia* / average algae	6.92	0.094	0.027	1.70	1.62	0.99	0.584	0.28	0.234	0.244	1.69
Average *Daphnia* / spring water	1264	4208	153	1936	5407	241	599	0.12	4.28	1.53	0.40
Study: Cowgill and Prance (1982)	**Ca**	**Mg**	**Be**	**Sr**	**Ba**	**B**	**Al**				
Victoria amazonica											
Young plants	4054	4107	0.064	35.7	105	15.6	4391	0.156	259	88.1	35.6
Pre-flowering plants	9508	4012	0.041	18.5	46.9	12.2	779	0.102	48.9	19.5	156
Mature plants, unopened blooms	8303	3703	0.037	14.9	50.2	15.6	365	0.10	60.2	17.9	427
Mature plants, full blooms	6421	3109	0.033	10.8	51.4	15.2	324	0.106	80.1	16.8	469
Study: Cowgill (1973*a*)											
Nymphaea odorata (water lily)	10,200	2500	0.445	16.7	42.3			1.78	16.3	41.5	
Rhopalosiphum nymphaea (aphid)	11,900	2300	0.32	25.5	82.2			1.39	21.4	69.1	
	Bioaccumulation factors										
Aphid/water lily	1.17	0.92	0.72	1.53	1.94						

A recent data set from Yan *et al.* (1989) provides uptake data for the net zooplankton for a series of lakes in the Canadian Shield. Since the Canadian Shield is a region currently under consideration by the Government of Canada as a possible site for a high-level nuclear waste repository, the biota of this region are of Task Force interest. Furthermore, the region is within one day's automobile travel of the Great Lakes, and depending on a variety of climate and other geophysical factors, the possibility of radionuclides in this region reaching the Great Lakes cannot be discounted. Further, and quite fortunately, most of biota found in the lakes studied also occur in the upper Great Lakes, making the zooplankton studies of these lakes relevant to an understanding of the biological cycling of elements in comparable biota in the Great Lakes. These very important data appear in Table 12.

The data suggest that strontium and barium have very different uptake characteristics relative to the Group II elements. Strontium uptake and levels do not appear to depend on the uptake and tissue levels of calcium, but barium uptake and levels show strong statistical correlations with calcium levels. The correlations between barium and calcium levels in zooplankton are particularly pronounced when the calcium level exceeds 10,000 ppm dry weight of tissue.

The separation of lakes into Sudbury and non-Sudbury area reflects the investigators' concerns about the discharges of nickel from the Sudbury smelter as a point source of pollution. This pollution source strongly influences the overall elemental uptake of metals by zooplankton in nearby lakes. Yan *et al.* (1989) chose lakes that were not themselves known to be polluted from other point sources or nonpoint sources, paying careful attention to obtaining a range of chemical conditions with respect to alkalinity, pH (a concern about lake acidity, which again is strongly influenced by smelter releases of sulfur oxide gases and aerosols), organic content, and diversity of zooplankton species. Thus non-Sudbury area lakes would not be expected to have zooplankton with high tissue levels of nickel unless atmospheric transport and deposition of nickel to those lakes occurred. The Task Force did not consider whether a calcium correlation with nickel influenced a calcium correlation with strontium or barium because of the inadequacy of the statistical tools available for such comparisons with these special data.

TABLE 12
ELEMENTAL COMPOSITION OF THE BIOTA FROM LAKES IN THE CANADIAN SHIELD. PART I — UPTAKE OF STRONTIUM, BARIUM AND RELATED ELEMENTS BY NET PLANKTON

(adapted from Yan *et al.* 1989)

	Elements (ppm)				Geochemical ratios ($\times 10^3$)	
Lake	**Ca**	**Mg**	**Sr**	**Ba**	**Sr/Ca**	**Ba/Ca**
Sudbury area:						
Clearwater	119	113	0.1	1.2	0.84	10.0
Lohi	816	864	3.0	4.4	3.68	5.39
McFarlane	109,635	4480	197.0	82.9	1.80	0.76
Tyson	5528	2487	35.1	7.0	6.35	1.27
Attlee	31,551	1705	159.0	185.0	5.04	5.86
Ruth Roy South	149	949	4.2	13.6	3.66	11.8
Ruth Roy North	836	888	3.9	14.5	4.67	17.3
Non-Sudbury area:						
Heney	1906	1258	12.3	26.4	6.45	13.9
Dickie	5259	1387	27.3	24.4	5.19	4.63
Blue Chalk	10,965	948	52.7	33.2	4.80	3.03
Chub	8451	1867	56.7	65.0	6.71	7.69
Echo	6177	2098	38.2	49.7	6.18	8.05
Beech	20,628	1995	107.0	50.2	5.18	2.43
Ril	6533	2128	39.8	37.1	6.09	5.68
Rock	10,427	1978	67.8	84.9	6.50	8.14
Whitefish	6606	1986	46.6	54.5	7.05	8.25
St.Peter	29,742	1429	120.0	75.0	4.04	2.52
Hay	15,229	1595	81.0	51.7	5.38	3.39
McKenzie	52,857	2053	227.0	153.0	4.29	2.89
Little Boulter	5003	1933	29.5	34.4	5.90	6.88
Red Chalk main	12,117	1087	61.5	44.3	5.08	3.66
Red Chalk east	5431	1501	35.7	50.4	6.57	9.28
Louisa	21,280	968	118.0	149.0	5.54	7.00
Whalley	23,680	1686	114.0	66.3	4.81	2.80
Cecebe	4107	1523	27.1	44.5	6.60	10.8
Horn-1	58,237	2098	290.0	193.0	4.98	3.31
Harp	5101	1818	37.2	36.6	7.29	7.17
Horn-2	52,470	12,383	248	233	4.73	4.44
Big Ohlmann	14,943	1590	44.1	29.7	2.95	1.98
Mackie	27,246	823	63.5	41.7	2.33	1.53
Dan's	61,675	1771	139	44.9	2.25	0.728
Maple	19,024	823	85.0	32.1	4.47	1.69
Hill's	8051	477	40.2	24.4	4.99	3.03
Fortune	59,409	1706	125	35.8	2.10	0.606
Lucky	16,285	836	55.5	13.5	3.40	0.829
Faun	3910	878	20.3	12.5	5.19	3.20
Brandy	30,558	878	244	106	7.98	3.47
Bat	730	1378	0.8	1.4	1.09	1.92

TABLE 12 (continued)
ELEMENTAL COMPOSITION OF THE BIOTA FROM A SERIES OF LAKES IN THE CANADIAN SHIELD — NET ZOOPLANKTON
(adapted from Yan *et al.* 1989)

Statistics:

Average geochemical ratio	**Sudbury Area Lakes**	**Non-Sudbury Area Lakes**	**All lakes**
Average Sr/Ca	3.72	4.86	4.65
Average Ba/Ca (for all lakes):	7.47	4.39	5.02
(Ca in zooplankton ≤10,000 ppm)	9.15	6.96	5.92
(Ca in zooplankton >10,000 ppm)	3.31	3.39	3.37

There does not appear to be any statistically significant correlation between the following geochemical ratios: Ba/Ca and Ca/Sr.

Notes: (1) All data are dry weight.
(2) Geochemical ratio statistics for " lakes regardless of Ca" ignores correlation with calcium. Averages are taken over all geochemical ratios for a given element.

Composition of lake zooplankton by percentage of total biomass:

Taxa	**Range**	**Median**
Chydoridae	0–2.6%	0
Bosminids (total) *Bosmina longirostris* *Eubosmina tubicen* *E. longispina*	0–86.8	1.45
Diaphanosoma spp.	0–18.9	0.3
Daphnia spp.	0–96.6	19.4
Holopedium gibberum	0–82.9	2.95
Predatory Cladocera *Polyphemus pediculus* *Leptodora kindtii*	0–6.82	0
Calanoida	0.14–100	25.5
Cyclopoida	0–72.6	8.51

GROUP IIb ELEMENTS: ZINC, CADMIUM, AND MERCURY

Zinc, cadmium, and mercury

The Group IIb elements, zinc, cadmium, and mercury, are important heavy metal contaminants of the Great Lakes. Each element has at least one radionuclide potentially requiring an inventory for the Great Lakes (^{65}Zn, ^{113}Cd, ^{203}Hg), but actually only the radionuclide of zinc needs a separate inventory for biological compartments.

^{65}Zn is an activation product with a long enough half-life (244 days) to cycle within the Great Lakes. Zinc is also a required trace metal in many metabolic processes, and thus, zinc will accumulate quite easily in biological tissues.

A nuclide of cadmium occurs naturally and the radioactive portion of this nuclide found naturally in biota can be obtained by a procedure similar to the one used for ^{40}K. Another nuclide of cadmium, ^{109}Cd (half-life 450 days), occasionally appears in discharges of nuclear facilities and is used in laboratory research (see discussions of the Task Force of Tier II nuclear facilities). Although its half-life assures its cycling through biological compartments, the source data on its discharge into the Great Lakes are inadequate to produce an estimated inventory.

The nuclide of mercury, although it is a fission product, has a very low fission yield and does not routinely occur in nuclear effluents or wastes. It has been used in laboratory and field research, but inadequate data preclude preparation of an estimate of its inventory in Great Lakes biotic compartments.

GROUP IIIa ELEMENTS: BORON, ALUMINUM, GALLIUM, INDIUM, AND THALLIUM

The Group IIIa elements are boron, aluminum, gallium, indium, and thallium. Of these elements, only boron and aluminum are of concern in developing inventories of radionuclides. Gallium, indium, and thallium do not have nuclides that are produced in nuclear systems that are likely to be discharged to the Great Lakes, although Cowgill has noted that gallium is accumulated by a variety of organisms.

Boron

Boron is a component of moderator materials in the reactors of nuclear power plants because of its high neutron absorption abilities. There are no reports of radionuclides of boron released to the Great Lakes. Rather the products of nuclear reactions of boron are nuclides of other elements, the most important of which is tritium. Boron is a plant nutrient, and in excess, a plant toxicant. Since few compounds of boron are water soluble, the range of concentrations of boron in source material exerting effects on organisms is quite small.

Aluminum

Aluminum is a reference element often chosen to describe geochemical coherence of elements originating in soils and minerals. Plants can accumulate aluminum, and an early study by Hutchinson and Wollack (1943) examined aluminum accumulator plants in considerable detail. Also, two radionuclides of aluminum, ^{26}Al and ^{28}Al, are produced cosmogenically. Data for aluminum may assist in the understanding of the behavior of other elements, but no separate bioaccumulation factors for aluminum are given.

GROUP IIIb ELEMENTS: SCANDIUM, YTTRIUM, LANTHANUM, AND RARE EARTHS

The Group IIIb elements of the Periodic Table are rather unusual. All are rare in nature. The low molecular weight scandium often shows up in particulate matter sampled in the upper atmosphere and has two nuclides which form cosmogenically. It has also been studied by Cowgill in plants, but the data on potential discharges to the Great Lakes are scant.

Yttrium and lanthanum and the other rare earth elements, are well represented among the nuclides formed in the nuclear fuel cycle. Many rare earth elements have primordial radionuclides among their isotopes, and almost all of the isotopes of the rare earth elements are mildly radioactive. The behavior of this group of elements in biological materials is not well understood. Cowgill's studies of the rare earth elements in various plant species suggested that organisms may exert considerable selectivity on which elements they accumulate, that they can accumulate considerable quantities relative to the low content of these elements in source materials, and that the elements can remain largely undetectable by present methods, suggesting levels below 1 ppm in either source materials or biological tissues.

Except for scandium, radionuclide inventories in biological compartments are needed for all of the Group IIIb elements and rare earths, although some scandium data are provided for reference purposes. The data are presented in Table 13.

Yttrium

Radionuclides of yttrium form directly as fission products and as the decay products of other fission products, notably radionuclides of Sr. Two nuclides of yttrium have been reported in discharges to the Great Lakes, ^{90}Y and ^{91}Y. The former has a very short half-life, but the latter has a sufficiently long half-life to call for an inventory. Yttrium can accumulate in organisms, but the available data base is sparse. Cowgill's data presents the most comprehensive review of yttrium in aquatic organisms. The environmental cycling of yttrium appears to follow that of the rare earth elements described below.

Lanthanum

Lanthanum is one of the most toxic elements to aquatic organisms. Nevertheless, it is detectable in small amounts in aquatic biota along with other rare earth elements.

Lanthanum phosphate is very insoluble in water. The precipitate is quantitative with a very high gravimetric constant, thus providing an analytical method for the element. This information suggests that lanthanum would not be expected to be a water pollution problem for most waters given their phosphate content. Further, the rarity of the element makes its recovery from aquatic systems financially worthwhile rather than discharging it in a waste effluent.

The accumulation of lanthanum in biota seems to depend on the presence of calcium. Lanthanum can sometimes substitute for calcium in a biouptake process from a calcium-impoverished medium. Since calcium phosphates are also insoluble, this may offer a partial explanation of the behavior of lanthanum.

One radionuclide of lanthanum important for Great Lakes work is ^{140}La. It forms directly as a fission product and as the decay product of ^{140}Ba. This latter fact explains why several dischargers tend to report the combination of ^{140}Ba/^{140}La , without separating the relative proportions of the isotopes.

Cerium

Cerium is only other rare earth element besides lanthanum that is routinely detected in biota. Cowgill's data probably provides the most complete set of information on the stable forms of the element in aquatic biota. Radionuclides of cerium are major fission products and have been detected in Great Lakes waters. The two major radionuclides of interest are ^{141}Ce and ^{144}Ce.

Other Lanthanides

As previously noted, organisms may be capable of accumulating rare earth elements selectively. Except for lanthanum and praeseodymium, Cowgill (1974*a*) found that the macrophytes of Linsley Pond only accumulated elements of even atomic number. She did not observe the pattern repeated in her studies of the giant Amazon River water lily, *Victoria amazonica*. For that species, the only detected rare earth elements were lanthanum and cerium (Table 14).

In her studies of the rare earth elements accumulated by *Daphnia*, Cowgill found europium in the organisms, but not in the source materials, suggesting that it was highly impoverished in the source materials composing the *Daphnia* medium, but that the organisms could still bioaccumulate and concentrate to a great degree. With respect to bioaccumulation processes in general, the lanthanides act sufficiently coherently that one can treat the total accumulation of all of those elements as a single element for bioaccumulation analyses.

In several studies the behavior of unusual elements that cycle in lakes and the oceans through sorption to the surfaces of bioparticles, a radionuclide of gadolinium was used. However, the authors of that study could not provide background data on gadolinium levels in the aquatic environment to assist in the interpretation of their work. It is not clear whether gadolinium should be considered an element which cycles on an organism as opposed to in an organism.

TABLE 13
UPTAKE OF RARE EARTH (RE) ELEMENTS AND THEIR CONGENERS BY AQUATIC BIOTA
(from Cowgill 1976)

Species or substrate	**Elements (ppm)**											
	Y	**La**	**Ce**	**Pr**	**Nd**	**Sm**	**Gd**	**Dy**	**Er**	**Yb**	**Other RE**	**∑RE**
Euglena gracilis	53.3	23.7	48.8	4.3	9.4	4.4	4.2	2.5	1.2	1.9	Eu 1.3	101.7
Mixed algae	61.5	22.2	43.6	2.5	7.8	4.0	3.7	1.8	1.3	1.7	Eu 1.4	90.0
Daphnia pulex (culture)	32.4	54.8	44.1	4.8	20.4	5.5	4.8	2.7	0.7	1.9	Eu 1.4	141.1
Daphnia magna (culture)	50.6	47.4	41.6	5.8	15.4	4.2	3.5	3.7	0.7	1.4	Eu 0.9	124.6
Average *Daphnia*	41.4	51.1	43.1	5.3	17.9	4.9	4.3	3.2	0.7	1.7	Eu 1.2	132.9
Trap rock	19.7	11.5	71.5	3.4	9.7	3.1	2.8	0.78	4.9	3.3	Eu 1.1	112.1
Spring water ($\times 10^{-5}$)	160	90	240	23	95	4.4	9.8	16	6	20	Eu nd	664
Bioaccumulation factors												
Euglena gracilis/spring water	33,312	26,333	20,333	18,695	9894	100,000	42,857	15,625	20,000	950		15,317
Mixed algae/spring water	38,438	24,667	18,167	10,870	8211	90,909	37,755	11,250	21,667	850		13,554
Daphnia pulex/spring water	20,250	60,889	18,375	20,870	21,477	125,000	48,980	16,875	11,667	950		21,250
Daphnia magna/spring water	31,625	56,222	17,333	25,217	16,210	107,955	43,368	23,125	11,667	700		18,765
Average *Daphnia*/spring water	25,875	58,556	17,854	23,044	18,844	116,478	46,174	20,000	11,667	825		20,008
Daphnia pulex/mixed algae	0.53	2.46	1.01	1.92	2.62	1.38	1.30	1.5	0.54	1.11	Eu 1.0	1.57
Daphnia magna/mixed algae	0.91	2.28	0.95	2.32	1.97	1.18	1.15	2.1	0.54	0.83	Eu 0.64	1.38
Average *Daphnia*/mixed algae	0.74	0.67	0.98	2.12	2.30	1.23	1.23	1.8	0.54	0.97	Eu 0.82	1.48

TABLE 13 (continued)
UPTAKE OF RARE EARTH ELEMENTS AND THEIR CONGENERS BY AQUATIC BIOTA
(from Cowgill 1976)

		Geochemical ratios					
Species or substrate	**∑RE/Ti**	**Ca/Ti**	**Ca/La**	**Ca/∑RE**	**P/Ti**	**P/La**	**P/∑RE**
Earth's crust		7.37	$>10^{18}$	$>2.8 \times 10^{9}$	0.184	34.8	
Trap rock	0.0823	8.36			0.060	3.53	0.73
Spring water	1.4805	1122			7.25	32.2	4.89
Euglena gracilis	8.92						
Mixed algae	6.66	570			1876	1180	282
Daphnia pulex	58.80						
Daphnia magna	54.18						
Average *Daphnia*	56.5	24,153			6186	285	109
		Ca/Ti	**Ca/La**	**Ca/∑RE**	**P/Ti**	**P/La**	**P/∑RE**
Earth's crust		7.37	1391		0.184	34.8	
Trap Rock		8.36	4918	2882	0.060	3.53	0.73
Spring Water		1122	4987	758	7.25	32.2	4.89
Euglena gracilis							
Mixed Algae		570	358	85.6	1876	1180	282
Daphnia pulex							
Daphnia magna							
Average *Daphnia*		24,153	1111	427	6186	285	109

TABLE 13 (continued)
UPTAKE OF RARE EARTH ELEMENTS AND CHEMICAL CONGENERS BY AQUATIC BIOTA:
Y, La, Ce, Pr, Nd, Sm, Gd, Dy, Er, Yb
(adapted from data of Cowgill)

	Elements (ppm)											
Species or substrate	**Y**	**La**	**Ce**	**Pr**	**Nd**	**Sm**	**Gd**	**Dy**	**Er**	**Yb**	**Other RE**	**$\sum$RE**
Nymphaea odorata (1971)												
Flowers	74.1	63.2	85.1	5.7	12.6	20.0	5.6	1.5	2.3	1.6		197.6
Flower stem	66.6	46.4	69.5	5.0	11.5	19.1	4.3	2.0	2.3	1.5		159.6
Leaves	82.2	32.6	93.4	5.8	12.8	22.0	2.9	4.1	4.6	1.7		179.9
Stem	65.6	39.5	76.0	3.5	17.6	19.4	2.7	1.2	2.8	1.5		164.2
Nymphaea odorata (1972)												
Flowers	70.1	57.1	111	6.1	11.6	19.9	5.4	3.6	5.4	1.5		221.3
Flower stem	62.7	46.1	98.3	4.3	9.6	18.8	4.5	3.4	5.8	1.4		
Leaves	72.9	27.4	111	3.8	15.1	19.4	5.4	3.4	4.4	1.5		
Stem	56.9	31.8	92.1	4.7	13.5	11.7	4.7	3.5	3.4	1.3		
Rhopalosyphum nymphaea	74.0	58.2	120.4	6.1	10.0	22.4	5.9	3.2	5.0	1.7		232.9
	Bioaccumulation factors											
Aphids/*N. odorata* leaves	0.90	1.79	1.29	1.61	0.78	1.02	2.03	0.78	1.09	1.0	1.29	
Aphids/*N. odorata* flowers	0.99	0.92	1.41	1.0	0.79	1.12	1.05	2.13	2.17			

TABLE 14
UPTAKE OF RARE EARTH ELEMENTS AND CHEMICAL CONGENERS BY AQUATIC BIOTA:
Y, La, Ce, Pr, Nd, Sm, Gd, Dy, Er, Yb
(adapted from data of Cowgill)

Species or substrate	Elements (ppm)											
	Y	**La**	**Ce**	**Pr**	**Nd**	**Sm**	**Gd**	**Dy**	**Er**	**Yb**	**Others**	**$\sum$RE**
Study: Cowgill and Prance (1982)												
Victoria amazonica (Amazon River)												
Young plants	0.208	0.92	0.74									
Pre-flowering plants	0.285	0.88	0.89									
Unopened bud plants	0.324	1.0	0.81									
Full blooming plants	0.328	0.93	0.68									

GROUP IVa ELEMENTS: CARBON, SILICON, GERMANIUM, TIN, AND LEAD

The Group IVa elements are carbon, silicon, germanium, tin, and lead. Although they share a common chemical grouping, their chemistry differs very markedly from element to element. For that reason, each element is discussed separately without any major prefatory statement. Data on uptake of Group IVa elements is presented in Table 15.

Carbon

The major radionuclide of carbon is ^{14}C, and it requires consideration all by itself. However, bioaccumulation factors for carbon would require a consideration of all the compounds which are formed. Thus, bioaccumulation factors are not developed for ^{14}C, per se, but specific inorganic and organic compounds that contain the element.

Silicon

Silicon is an essential element in the skeletal structure of diatoms and Foraminafera, usually in the amorphous mineralogical form of opaline phytoliths. The bioaccumulation of silica suggests that it might be a macronutrient in plants other than diatoms, but as earlier indicated, many silica analyses may be faulty because of analyses of silica on the material rather than silica in the material.

The biological cycling of silicon in the Great Lakes (because of diatoms) has been intensively studied by Stoermer and his coworkers. Wahlgren et al. (1980) used the cycling of silica to explain much of the dynamics of the movement of transuranics, especially plutonium, in Lake Michigan, particularly the movement of plutonium to sediments and seasonal distributions. The silicon–plutonium connection thus has particular importance in establishing inventories of transuranic radionuclides to the Great Lakes.

Two radionuclides of silicon, ^{31}Si and ^{32}Si, are produced cosmogenically. There is some limited production by fission, but no indication that the fission isotopes are released to the Great Lakes. The same procedures used to develop biocompartment inventories for the cosmogenically produced nuclides of aluminum can be used to produce biocompartment inventories for the radionuclides of silicon.

Germanium

Germanium does accumulate in biota, but there are many problems with analytical procedures and other technical concerns that mitigate making the most cursory estimates of germanium bioaccumulation factors. Germanium has not been detected or reported on in Great Lakes water or biota, but its presence is sufficiently difficult to measure that it might be overlooked. Germanium isotopes are found in fuel elements and therefore of possible concern, although as previously noted, the discharge of germanium to the Great Lakes is largely undocumented.

The analytical procedures for germanium are quite difficult, and there are many technical problems in detecting and quantifying this element in aquatic media. Some of these methodological difficulties cast doubt on the confidence of some of the data in the literature. Given the very limited data available for the element, there seems little justification in attempting what would be a highly speculative estimate of an inventory for germanium for biocompartments.

Tin

Tin is also a very difficult element of Group IV to consider. Very few data exist on its occurrence in aquatic systems and even less in biological compartments. Analytical methods to quantify tin in environmental media and substrates call for considerable skill and instrumental sophistication.

Two radionuclides of tin, ^{113}Sn (half-life 115 days) and ^{117m}Sn (half-life 14 days), occasionally are reported in the liquid emissions to the Great Lakes. Both nuclides have sufficiently long half-lives to undergo cycling through Great Lakes compartments. They also exhibit unusual decay patterns: ^{113}Sn decays by orbital electron capture to give a stable nuclide of indium, while ^{117m}Sn decays by an internal transition that produces a stable nuclide of tin. Thus, at least one tin nuclide continues to cycle environmentally unchanged. The nuclides originate from fission and as activation products of the zircalloy (a zirconium–tin alloy) sheathing for nuclear fuel elements, making them important nuclides from the nuclear fuel cycle.

Tin has a rather unusual chemistry and a very interesting toxicology. It forms several highly toxic metalorganic compounds (*i.e.*, tributyl tin). The methylation property permits biological mobilization of otherwise relatively water-insoluble tin compounds in the environment, permitting them to cross biological membranes and deposit in lipid tissue reserves. Further, selected organotin compounds are volatile.

Cowgill's data probably comprise the most extensive set of tin analyses on aquatic biota for nonradioactive materials, but her data base is small.

Lead

Isotopes of lead are the final products from the decay of transuranics. The long life of ^{210}Pb and the concern about lead as a toxicant and air pollutant have led researchers to study this element and this particular radionuclide intensively. Thus the information base on radioactive and stable lead is rather large, but the biological data on the radionuclide form of lead is not very extensive.

TABLE 15
UPTAKE OF GROUP IV, V, AND VI ELEMENTS BY AQUATIC BIOTA:
Si, Ge, Sn, Pb, S , Se, P (ppm)

Species or substrate	Si	Ge	Sn	Pb	S	Se	P
Study: Cowgill (1976)							
Euglena gracilis	725	0.25	23.5	12.6	5943	2.0	23095
Mixed algae	1136	0.18	20.0	10.0	4201	1.9	30960
Daphnia pulex	893	0.29	4.2	9.8	3965	2.1	13141
Daphnia magna	771	0.20	1.2	7.2	5647	1.6	15934
Study: Cowgill (1974*a*)							
Nymphaea odorata (Linsley Pond, 1971)							
Flowers	11,800	0.34	5.0	11.0			
Flower stalks	3600	0.30	7.5	10.7			
Leaves	10,600	0.34	3.2	12.0			
Stems	1800	0.27	4.4	9.6			
Study: Cowgill (1973*a*, 1974*a*)							
Nymphaea odorata (Linsley Pond, 1972)							
Flowers	680	0.36	3.0	9.8			
Flower stalks	640	0.31	9.0	9.6			
Leaves	570	0.33	6.4	8.4			
Stems	620	0.27	10.0	9.0			
Nymphaea odorata (Cedar Lake, 1971)							
Flowers	14,600	0.37	2.8	11.5			
Flower stalks	2800	0.30	7.1	11.9			
Leaves	9000	0.33	3.4	12.6			
Stems	4800	0.27	4.2	11.7			
Rhopalisyphum nymphaea	1760	0.55	2.5	9.2			
Study: Uthe and Bligh (1971)			Special note: wet weight data; estimated moisture content 80%				
Coregonus clupeaformis							
Moose Lake, Manitoba			3.57	<0.5	0.24		
Lake Ontario			0.80		0.38		
Esox lucius							
Moose Lake, Manitoba			5.43		0.17		
Lake St. Pierre			0.67		0.37		
Lake Erie			0.54		0.19		

GROUPS IVb AND Vb ELEMENTS: TITANIUM, ZIRCONIUM, HAFNIUM, VANADIUM, ANTIMONY, AND TANTALUM

The Group IVb and Group Vb elements include titanium, zirconium, hafnium, vanadium, antimony, and tantalum. Other than for antimony and tantalum, for which biological data are either exceedingly rare or nonexistent, the other elements in these groups present an interesting combination with respect to bioaccumulation. Uptake data for some of these elements are presented in Table 16.

Titanium

Titanium is among the least biological mobile of the elements and, thus, is a favorite choice of geochemists as a reference element for studies of elements that move preferentially with soil or mineral materials rather than with biological materials. Nevertheless, titanium accumulates in organisms, and some recent data from Yan *et al.* (1989) on a lake in the Canadian Shield region show an unusually enriched titanium level in zooplankton. The great insolubility of titanium in water can sometimes suggest that high titanium levels found in organisms may possibly originate as external titanium contamination of materials (titanium on rather than titanium in biological tissues) or the ingestion of particles that become trapped in some storage matrix in the alimentary canal of organisms without mobilization to other tissues. On the other hand, titanium citrate is soluble in aqueous solution at the levels of titanium detected in biota and the water column, and the presence of citrate as a major organic compound in all organismal tissue offers an alternative hypothesis to explain the apparently anomalous titanium data. The data from titanium analyses in freshwater biota do not indicate whether or not titanium in biological tissues is trapped in a storage matrix and thus biologically inert.

There are no indications that radionuclides of titanium are discharged to the Great Lakes, although several are produced as fission products and would be included in the inventory of nuclides for used fuel elements. Bioaccumulation data for titanium are used to assist in estimation of properties of other elements.

Zirconium and niobium

Zirconium and niobium are metals associated with the preparation of shielding material to contain fuel elements in nuclear power plants. Their isotopes form by both neutron activation and fission products. Atmospheric testing of nuclear weapons prior to 1963 produced zirconium nuclides, which were detected in atmospheric fallout. Radionuclides of zirconium decay to radionuclides of niobium, and the specific combination of ^{95}Zr/^{95}Nb is sometimes treated as one nuclide in the emission reports of nuclear facilities (the reader should recall the situation with ^{140}Ba/^{140}La). Both ^{95}Zr (half-life 65 days) and ^{95}Nb (half-life 35 days) have sufficiently long half-lives to be of environmental concern. The radionuclide, ^{94}Nb (half-life 20,000 years), is one of the longest lived, but there are no indications that it is discharged directly to the Great Lakes region. Both elements bioaccumulate in tissues. In the few available studies, niobium accumulates to a greater extent than zirconium, at least in plants. The available data for each element are quite limited, with Cowgill's work probably the most comprehensive source. Cowgill evaluated her niobium data relative to vanadium. This offers a useful statistical approach because the availability of biological data for vanadium far exceeds that of niobium. The methodology studies Nb/V ratios as guides to estimating niobium inventories in unstudied situations.

Hafnium

Hafnium has several long-lived isotopes, including one that is primordial. It also accumulates to a very limited degree in plants. The limited data come mainly from Cowgill.

Vanadium

Vanadium accumulates in organisms (Table 16) and is an essential micronutrient for certain plants and fungi. ^{50}V is a primordial radionuclide. Other radionuclides of vanadium form as both fission and activation products. Recent interest in vanadium by a number of researchers has provided a data base on vanadium uptake in plants and animals that offers possibilities in estimating its behavior in biological compartments. However, there is only limited indication that vanadium radionuclides are released to the Great Lakes. Rather, the importance of vanadium data assist in studying niobium.

TABLE 16
UPTAKE OF SELECTED TRANSITION ELEMENTS BY AQUATIC BIOTA:
Ti, V, Cr, Mn, Fe, Co, Ni, Zn, Zr, Nb, Mo
(adapted from data of various investigators)

	Elements (ppm)										
Species or substrate	**Ti**	**V**	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Zn**	**Zr**	**Nb**	**Mo**
Study: Cowgill (1976)											
Euglena gracilis	11.4	6.4	1.4	279	12,961	3.8	1.8	304	7.6	14.1	6.5
Mixed algal culture	17.4	2.8	1.2	240	10,534	2.8	2.7	339	9.8	13.3	5.7
Study: Cowgill (1973*a*)											
Pontaderia cordata											
(Linsley Pond)											
Stem	11.1	6.3				0.10	3.9			14.2	
Leaf	10.9	80.0				1.7	4.0			13.4	
Flower	9.1	10.3				0.10	2.8			14.8	
Pontaderia cordata											
(Cedar Lake)											
Stem	48.7	8.0				0.14	2.7		30.7	13.4	
Leaf	13.2	79.0				4.6	3.9		41.5	11.8	
Flower	12.4	9.0				0.06	2.9		22.8	13.4	
Nuphar advena											
(Linsley Pond)											
Leaf petiole	20.3					0.04	3.1		32.1		
Leaf	13.8					0.11	3.4		41.5		
Flower stalk	12.8					0.08	2.9		28.7		
Flower	13.0					0.09	4.5		31.1		
Nymphaea odorata											
(Linsley Pond, 1971)											
Flowers	8.0		0.55	151	5100	0.13	3.1	137	20.4		0.08
Flower stem	17.0		0.50	191	7800	0.05	2.7	128	18.2		0.27
Leaves	11.5		0.64	245	9200	0.08	2.8	134	41.9		0.07
Stem	13.4		0.52	233	3600	0.07	2.4	111	19.1		0.43

TABLE 16 (continued)
UPTAKE OF SELECTED TRANSITION ELEMENTS BY AQUATIC BIOTA:
Ti, V, Cr, Mn, Fe, Co, Ni, Zn, Zr, Nb, Mo
(adapted from data of various investigators)

Species or substrate				**Elements (ppm)**							
	Ti	**V**	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Zn**	**Zr**	**Nb**	**Mo**
Nuphar advena (Linsley Pond ,1972)											
Flowers	2.7		0.64	247	1040	0.25	4.2	105	8.7		0.05
Flower stem	2.6		0.53	151	1320	0.19	3.4	110	13.1		0.18
Leaves	2.5		0.60	719	1290	0.22	4.8	139	10.1		0.10
Stem	3.1		0.64	492	1100	0.19	2.7	110	12.0		0.30
Study: Cowgill and Prance (1982)											
Victoria amazonica (Amazon River)											
Young plants	20.1	0.076	2.23	3807	8929	0.23	0.40	15.2	5.9	0.29	0.315
Pre-flowering plants	3.3	0.067	2.05	6532	2835	0.27	0.49	19.0	2.2	0.34	0.330
Unopened bud plants	1.3	0.063	2.0	3242	1349	0.73	0.55	19.7	1.5	0.38	0.289
Full blooming plants	0.9	0.056	1.9	2417	1034	0.20	0.59	20.9	1.2	0.39	0.250
Study: Steinnes (1995)											
Holocomium splendens (Svalbard, Sweden)											
Station I		3.9	2.5		1176		3.7				
Station II		5.2	3.6		2076		2.6				
Station III		17.4	9.1		4501		5.2				
Station IV		2.9	1.5		1420		1.3				
(Iceland)											
Station I		10.0	5.2		4059		5.7				
Station II		15.3	2.8		4952		3.2				
Station III		8.0	3.4		2782		3.3				
Study: Cowgill (1976)											
Daphnia pulex	2.4	4.4	1.3	132	1014	3.3	4.2	135	14.9	16.1	0.7
Daphnia magna	2.3	1.3	0.6	94	916	0.7	3.6	102	15.9	11.9	0.2
Average *Daphnia*	2.3	2.9	1.0	113	965	2.0	3.9	119	15.4	14.0	0.5
Study: Cowgill (1974*a*)											
Rhopalosiphum nymphaea	28.6		0.88	780	3070	0.5	5.3	152	8.7		2.0

TABLE 16 (continued)
UPTAKE OF SELECTED TRANSITION ELEMENTS BY AQUATIC BIOTA: Ti, V, Cr, Mn, Fe, Co, Ni, Zn, Zr, Nb, Mo
(adapted from data of various investigators)

Species or substrate	**Elements (ppm)**										
	Ti	**V**	**Cr**	**Mn**	**Fe**	**Co**	**Ni**	**Zn**	**Zr**	**Nb**	**Mo**
Study: Yan *et al.* (1989)											
Net zooplankton in selected lakes											
Heney	2.85			36.9	897		3.95	112			
Dickie	5.2			88.5	1075		0.30	95			
Blue Chalk	4.6			138	586		0.18	85			
Rock	15.4			243	1942		0.10	126			
McKenzie	61.0			454	3229		1.80	135			
Whalley	11.0			186	1686		3.06	92			
Horn-1	73.0			303	2474		7.43	94			
Horn-2	134			360	7458		2.09	1160			
Fortune	4.7			189	350		5.83	132			
Brandy	35.2			299	1842		8.90	101			
Bat	0.46			5.4	114		0.34	80			
Attlee	136			388	3262		21.6	123			
McFarlane	8.8			690	1089		329	368			

TABLE 16 (continued)
UPTAKE OF SELECTED TRANSITION ELEMENTS BY AQUATIC BIOTA:
Ti, V, Cr, Mn, Fe, Co, Ni, Zn, Zr, Nb, Mo
(adapted from data of various investigators)

	Geochemical ratios							
Study and species or substrate	**Ti/ V**	**Ti/Zr**	**Zr/Nb**	**V/Nb**	**Fe/Mn**	**Fe/Co**	**Fe/Ni**	**Cr/Mo**
Study: Cowgill (1976)								
Euglena gracilis	1.78	1.5	0.54	0.45	46.46	3410.79	7200.56	0.22
Mixed algae	6.21	1.78	0.74	0.21	43.89	3762.14	3901.48	0.21
Daphnia pulex	0.54	0.16	0.93	0.27	7.68	307.27	241.43	1.86
Daphnia magna	1.77	0.14	1.34	0.11	9.74	1308.57	254.44	3.00
Average *Daphnia*	0.79	0.15	1.10	0.21	8.54	482.5	247.44	2.00
Spring water								
Trap rock								
Study: Cowgill and Prance (1982)								
Victoria amazonica								
Young plants	265	3.41	203	0.26	2.34	38,821		
Pre-flowering plants	136	1.5	6.5	0.20	0.43	12,326		
Mature plants: unopened buds	21	0.87	39.4	0.17				

TABLE 16 (continued)
UPTAKE OF SELECTED TRANSITION ELEMENTS BY MAMMALS:
Ti, V, Cr, Mn, Fe, Co, Ni, Zn, Zr, Nb, Mo
(adapted from data of various investigators)

Species or substrate	Elements (ppm)										
	Ti	V	Cr	Mn	Fe	Co	Ni	Zn	Zr	Nb	Mo
Study: Elkin and Bethke (1995)											
Rangifer tarandus											
(Bathurst Station, NWT Canada)											
Kidney tissue			1.15	8.96	237		0.44	124			
Liver tissue			0.68	12.6	1594		0.45	114			
(Arviat Station, NWT Canada)											
Kidney tissue			1.67	12.0	218		0.24	121			
Liver tissue			0.90	10.8	702		0.49	93			
(Cape Dorset, NWT)											
Kidney tissue			1.23	11.7	441		0.90	107			
Liver tissue			0.62	8.6	3628		0.45	76			
(Lake Harbour, NWT)											
Kidney tissue			2.06	18.6	343		1.33	97			
Liver tissue			0.40	15.9	3956		0.13	76			
Study: Poole *et al.* (1995)											
Mustela vison (kidney tissue)											
(Northwest Territories, Canada)											
Inuvik 1992			0.49	3.7	814	1.18	76				
Inuvik 1993			1.33	2.3	840	0.48	82				
Fort Good Hope			0.45	11.2	965	1.89	68				
Fort Rae			0.44	5.0	958	1.32	104				
Fort Liard			1.09	4.0	966	0.61	122				
Fort Smith			1.17	2.2	852	0.45	84				

Notes: See page 73 and 75 for discussion of iron (Fe) and nickel (Ni) levels, respectively.

GROUP VIIa ELEMENTS (THE HALOGENS): FLUORINE, CHLORINE, BROMINE, IODINE, AND ASTATINE

The halogens, Group VIIa, of the Periodic Table are elements with univalent negatively charged ions: fluorine, chlorine, bromine, iodine, and astatine. Since astatine is an artificial nuclide, it is not discussed here. The remaining halogen elements exist naturally as diatomic gases or as salts in various minerals. The largest source fluorine is in minerals, especially certain aluminum and phosphate minerals. Chlorine, bromine, and iodine occur both as rock salt minerals as well as dissolved in ocean waters.

Fluorine

Fluorine has a very limited biological uptake. The carbon–fluorine bond is very strong, but it is also very difficult to create. A group of fluorine accumulator plants (terrestrial, and mainly found in Africa) apparently can metabolize and store fluorine through a fluoracetic acid pathway. Fluoracetic acid is very toxic to almost all animal species (terrestrial and aquatic) because it interferes with the Krebs' cycle. Radionuclides of fluorine have virtually no role in the Great Lakes, and there are no inventory calculations for fluorine presented.

Chlorine

Chlorine has several radionuclides, some of which are formed cosmogenically. Also chlorine is a macroelement in biological tissues because of its role in the maintenance of electrolyte balance and osmotic pressure, but studies of radioactive chlorine behavior in environmental systems are almost nonexistent. A review group evaluating the Draft Environmental Impact Assessment for high level waste deposit in a possible Canadian Shield repository noted with some surprise the apparently unexpected presence of detectable ^{36}Cl in reactor material. Although the nuclide can form as a fission product as well as cosmogenically, apparently its fission yield is so small that its appearance in the reactor materials was unexpected. The usual source of this nuclide is the decay of the cosmogenically produced ^{35}S. Consequently, some fraction of biologically retained chlorine would be expected to be radioactive.

Bromine

Bromine accumulates in several organisms and exhibits some unusual biological properties. It is a major inorganic constituent of the neurotoxins produced by several species of marine animals. It has been detected enriched in tumor tissue of a primate (Cowgill 1977), and the sodium or potassium bromide salts have long been used by humans for the treatment of headaches. Radionuclides of bromine form directly as fission products and as a decay product of other fission products. Although elemental bromine is a liquid at room temperature, it vaporizes easily and might accompany chlorine and iodine in gaseous effluents. However, most bromine occurs naturally as anionic material, usually either as bromide ion or bromate ion. Since most bromide and bromate compounds are very water soluble, the element attains a special importance should some accidental breach of nuclear fuel element integrity occur. Bromine would carry other nuclides directly into solution creating a high-level liquid radioactive waste. However, the radionuclides of bromine are very short lived.

Iodine

Iodine is the heaviest natural element of the halogen family, Group VII of the Periodic Table. Its congener elements, fluorine, chlorine, and bromine, have relatively few isotopes of biological significance and accumulation and are thus discussed together in a separate section of this chapter.

Freshwater and marine biota both accumulate iodine, but environmental levels of iodine are greatest in the oceans (Table 17). Thus marine plants, invertebrates, and vertebrates accumulate it to levels greater than freshwater or terrestrial organisms. The low levels of iodine in freshwater make its uptake almost uniquely limited to plants and vertebrates. There are very limited data on the uptake of iodine by freshwater invertebrates.

The biological role of iodine is unclear except in the vertebrates, where it is unique in thyroid gland function. Since the thyroid is a target organ in humans for radioactive iodine, both United States and Canadian regulations call for monitoring at least of the ^{131}I. This isotope is found in gaseous and sometimes liquid emissions from nuclear power plants. A much longer lived isotope, ^{129}I, occurs in the effluents of nuclear fuel reprocessing operations.

GROUP VIa ELEMENTS: SULFUR, SELENIUM, TELLURIUM, AND POLONIUM

The Group VIa elements are not considered. This may surprise people since isotopes of sulfur form by cosmogenic processes; ^{75}Se is a popular radioactive tracer of organismal metabolism for selenium; both sulfur and selenium are essential elements in the nutrition of all species, although selenium in excess is very toxic; tellurium is also quite toxic; and polonium is a radioactive product of uranium decay.

Cosmogenically produced nuclides of sulfur do not have long-term bioaccumulation potential. If they decay to stable sulfur, the formation of sulfate has implications for many environmental processes, but bioaccumulation and biomagnification are not among them. ^{75}Se is neither a fission nor activation product. All radionuclides of selenium produced by nuclear activities in the Great Lakes Region except for ^{79}Se are very short lived, but the residuals of ^{79}Se are important only in considerations of the high-level waste inventories for fuel elements. The Task Force has no documentation that selenium nuclides are released by nuclear facilities to the Great Lakes Basin. Those users of ^{75}Se for research purposes do not release that large a quantity that the cycling of this nuclide need be routinely considered; however, both sulfur and selenium are treated as elements that have importance in establishing the inventories and behaviors for the nuclides of other elements.

Despite the toxicity of tellurium, there is limited accumulation. Cowgill only found the element occurring naturally in plant material from the Middle East, not from her studies in North America (personal communication, April 26, 1996). Nor are radionuclides of tellurium documented in the releases from nuclear power plants, although such nuclides would require consideration in the high level waste inventories for fuel elements. Several isotopes of tellurium form as fission products. Many tellurium compounds are volatile and would appear in the gaseous emissions. Isotopes of tellurium have been documented in the gaseous emissions of fuel reprocessing operations, but not in North America.

TABLE 17
BIOACCUMULATION FACTORS AND BIOMAGNIFICATION FACTORS FOR VARIOUS ELEMENTS IN AQUATIC BIOTA

	Bioaccumulation				Biomagnification		
Elements	**Plants/ water**	**Invertebrates/ water**	**Fishes/ water**	**Plants/ sediments**	**Plants/ invertebrates**	**Fishes/ plants**	**Fishes/ invertebrates**
Beryllium	3000	—	—	—	0.05	—	—
Phosphorus	NA	NA	NA	NA	NA	NA	NA
Potassium	1000–3000	—	800	—	1–2	—	—
Chromium	50,000	50,000	—	—	1	—	—
Manganese	50,000	—	—	—	0.5	—	—
Iron	50,000	5000	—	—	0.1	—	—
Cobalt	300,000	60,000	—	—	1	—	—
Rubidium	10,000	12,000	—	—	1	—	—
Strontium	1000	—	—	—	1.5	—	—
Zirconium	2000	2000	—	—	1.5	—	—
Niobium	20,000	20,000	—	—	1.5	—	—
Iodine	2000	1000	—	—	0.4	—	—
Cesium	20,000	10,000	50	—	0.5	—	—
Cerium	20,000	—	—	—	1	—	—

Notes: NA, not available.
(—) The Task Force was unable to find data.

GROUP VIb ELEMENTS: CHROMIUM, MOLYBDENUM, AND TUNGSTEN

The group VIIb elements are chromium, molybdenum, and tungsten. All three elements have radionuclides formed by activation or fission processes that occur in the discharges to the Great Lakes. All three elements can bioaccumulate in tissues (Table 17).

Chromium

Chromium cycles through biological compartments in several valence states, two of which are very important in aquatic systems. The hexavalent (+6) state is very water soluble and highly toxic to most organisms. The trivalent (+3) state has a much lower water solubility and behaves as a trace micronutrient in certain tissue and organismal systems (Mertz 1967). Most environmental studies do not report the valence state of chromium, rather a "total" chromium level. As a precaution in toxicological and ecological studies, if the environment is aerobic, many investigators and most regulatory agencies assume that the "total chromium" is in the hexavalent state. Other valence states (+4, +10) of chromium are not stable (they can be explosive) and readily attack organic matter, especially under acid conditions.

Neutron activation produces an important radionuclide of chromium, ^{51}Cr. This nuclide behaves as a tracer of the effluent from nuclear power plants and scientists at the Hanford facility and Battelle Northwest have followed the nuclide's migration along the Columbia River to the Pacific Ocean for many years.

Molybdenum

Molybdenum is a trace nutrient requirement for plants, and functions in several enzymes associated with nitrogen fixation and the utilization of iron and sulfur in cellular metabolism. Two nuclides, ^{95}Mo and ^{99}Mo, form as fission and activation products. Although ^{99}Mo has a half-life of only 6 hours, it decays to ^{99}Tc (half-life 212,000 years) and thus plays an important role in estimating inventories for technetium. ^{99}Mo and ^{99}Tc are sometimes treated as a combined pair, similar to the treatment of ^{140}Ba and ^{140}La.

Tungsten

Tungsten has not been routinely detected in freshwater organisms. Most of the studies on biogeochemical cycling of tungsten examines the two radionuclides ^{181}W and ^{185}W, which are fission and activation products in nuclear reactors. These biological studies emphasized obtaining data to predict the exposure by humans to these nuclides through food. The studies emphasized tungsten uptake by terrestrial plants, notably agricultural crops. The data base upon which to make biological judgments about tungsten, in general and in the Great Lakes specifically, is very thin.

GROUP VIIb ELEMENTS: MANGANESE, TECHNETIUM, RHENIUM, IRON, RUTHENIUM, AND OSMIUM

The elements of VIIb come in two groups: (1) manganese, technetium, and rhenium and (2) iron, ruthenium, and osmium. Only manganese in the first subgroup and iron and ruthenium in the second subgroup have important nuclides for which inventories in biological compartments of the Great Lakes are needed. Technetium also requires study, but there is insufficient data and information to estimate its biological inventory in the Great Lakes. Rhenium and osmium, although highly toxic to mammals, are not documented in the discharges to the Great Lakes either directly as radionuclides or stable isotopes. Radionuclides of rhenium, decay products of radionuclides of ruthenium, have very short half-lives.

Iron and manganese

Iron and manganese are elements subject to the caution about "material on a tissue" versus "material in a tissue." Depending on the pH of the freshwater system or the "local pH" on the surface of tissue (plants that may have dew on leaves), and the level of oxygenation of the system (aerobic versus anaerobic environment), iron and manganese hydroxides can form precipitates on the external surfaces of biological materials. Many analyses of plants that report very high iron and manganese levels may actually have reported crystalline ferric and manganese oxides as surface contaminants. Despite these chemical artifacts, both elements have major metabolic roles in organisms and approach a status of macronutrient rather than simple trace nutritional requirement. Iron and manganese atoms occur in many enzymes associated with the metabolism of carbohydrates and lipids, as well as the production of proteins and nucleotides (DNA).

Radionuclides of iron and manganese form as activation products through neutron interactions with the piping and contaminants in the construction materials of fuel elements, reactor housings, etc. In addition, the casings on those nuclear weapons which were atmospherically detonated in the 1950s and early 1960s became activated with the release of radionuclides of these elements. Important nuclides are ^{55}Fe (half-life 2.6 years), ^{59}Fe (half-life 45 days), ^{54}Mn (half-life 303 days), and ^{56}Mn (half-life 2.6 hours). The two nuclides of iron and lower atomic weight nuclide of manganese last long enough to cycle through Great Lakes ecosystems. ^{55}Fe and ^{54}Mn decay by electron capture mechanisms, and ^{59}Fe and ^{56}Mn decay by β emission. The decay products of ^{56}Mn, a stable nuclide of iron, is itself subject of neutron activation. The remaining possible decay products undergo further decay by $\beta-$ and $\beta+$ emissions, further electron capture mechanisms, or some combination thereof. All daughter nuclides from these decay schemes have relatively high thermal neutron capture probabilities (*i.e.*, "cross sections").

The most important biological data on the radionuclides of iron and manganese come from studies of the marine environment and the monitoring of the Hanford facility discharges to the Columbia River and Pacific Ocean.

Ruthenium

Without nuclear technology, there would probably be very little interest in this element. Two radionuclides of ruthenium are produced in significant quantities in nuclear fuel processing and fission processes: ^{103}Ru (half-life 41 days) and ^{106}Ru (half-life 1 year). Both nuclides have sufficient time under a continuing discharge pattern to cycle within the biota of the Great Lakes, but the element has rarely been detected analytically in freshwater biota and reported in the literature.

The Radiation Protection Branch of the New York State Department of Health (1983–1993) has provided something of an exception to the monitoring of ruthenium. This group uses gamma radiation scintillation spectra to scan for selected radionuclides in biological materials collected from several sites in the New York portion of the Great Lakes Region. The problem, however, is that data reported for ^{106}Ru in biological samples show that the nuclide occurred at the lower limit of detection, if at all, and because the lower limit of detection differed from sample to sample, a range of lower limits of detection was observed. This makes inventory calculations very uncertain for ruthenium nuclides. Data for the Springville Dam site on Cattaraugus Creek (the site for monitoring effluents from the West Valley nuclear reprocessing facility that feed into Lake Erie) and Oswego, New York, near the Nine Mile Point nuclear power plant (Lake Ontario) appear in Table 18 and illustrate these issues.

Although ruthenium belongs to the same chemical family as iron and manganese, the Task Force considers it rather unwise to assume that the biological cycling of ruthenium follows the pattern of the biological cycling of iron and manganese. Available studies suggest that ruthenium moves to sediments and that most of the radiation it contributes to ecosystem burdens is gamma radiation from external exposure. The element has no known biological role. The Task Force has chosen not to estimate a biocompartment inventory for this element given its lack of observation in major studies of the elemental composition of freshwater biota.

TABLE 18
RADIONUCLIDES IN THE GREAT LAKES REGION: ^{106}Ru, ^{137}Cs, and ^{40}K
(adapted from the Annual Reports of the
Radiation Protection Branch of the New York State Department of Health 1983–1993)

	Cattaraugus Creek/Springville Dam					
Year	Water (pCi/L)		Fish (pCi/kg)			
	^{106}Ru	^{137}Cs	^{106}Ru	^{137}Cs	^{40}K	^{137}Cs/^{40}K
1986	<30–40	<6.0–8	<60	26 ± 20	2600 ± 200 (2)	0.010
1987	*	*	<30–60	<15–20	2850 ± 500 (2)	
1988	<30–50	<8.0–11	<50	27 ± 15	2300 ± 300 (3)	0.011
1989	<40–60	<10–12	*	*	*	
1990	<40–70	<10–13	<70–80	<12–15	2500 ± 300 (3)	
1991	<20–200	<6.0–40	<80–130	<12–20	3220 ± 460 (4)	
1992	<20–30	<6.0–8	<30–60	<7–15	2700 ± 245 (4)	
1993	<19–40	<5.0–10	<70–160	25 ± 18	2730 ± 530 (3)	0.0092

Notes: (1) Fish data are wet weight.
(2) Symbols: asterisk (*) means data not reported; the "less than" (<) means at or below level of detection with the first number being the level of detection for the samples considered; ± symbols indicate either the reported value with uncertainty or standard error or a mean value of several entries with a mean of standard error. In the latter case the number in parenthesis indicates the number of entries used in averaging (applies mainly to potassium data).

Rhenium and Osmium

What was said of ruthenium generally applies to rhenium and osmium. The radionuclides, except for a primordial nuclide of rhenium (^{187}Re), are very short lived. The elements have no known biological role but are toxic. They are rare in the earth's crust, and despite their toxicity, they are usually not detected in freshwater biota.

Technetium

This artificial element has several isotopes, all radioactive, but the most important isotope is ^{99}Tc (half-life 212,000 years). The remaining isotopes are short lived and not considered. Technetium bioaccumulates in tissues, but its pathways of biological cycling are almost unknown. There have been no comprehensive environmental studies, although its expected chemical form of technetium in aqueous systems is the TcO_4^{-1}, most of whose compounds are soluble. This makes the element potentially available for uptake by organisms in the water column.

Radionuclides of technetium result from fission and neutron activation. The isotopes also have use as medical diagnostic agents. Thus, there are many sources of technetium to the Great Lakes, but without a considerable quantity of fundamental data on water levels and biotic levels, no bioaccumulation factors are estimatable.

GROUP VIIIB ELEMENTS: COBALT, RHODIUM, IRIDIUM, NICKEL, PALLADIUM, AND PLATINUM

Similar to the Group VIIb elements, the Group VIIIb elements also come in two subgroupings: (1) cobalt, rhodium, and iridium and (2) nickel, palladium, and platinum. Only cobalt from the first subgrouping and nickel from the second subgrouping have radionuclides that are potentially bioaccumulation problems for Great Lakes biota (Table 17). The radionuclides of both elements are activation products and among those for which environmental monitoring is called for if detected.

Cobalt

All radionuclides of cobalt form as activation products. Four radionuclides, ^{57}Co, ^{58}Co, ^{59}Co, and ^{60}Co are discharged to the Great Lakes. Two of the nuclides, ^{60}Co (half-life 5.26 years) and ^{57}Co (half-life 270 days) are important in the discharges to the Great Lakes. The former has commercial use, and the latter is a major nuclide in research.

Cobalt is an essential micronutrient and is the metallic element that activates vitamin B_{12}. Therefore, cobalt biouptake is expected in all aquatic biota. Since a radioactive version of the vitamin is a source of internal radiation to blood forming organs and the liver in vertebrates, the cycling of cobalt is of both health and ecological interest.

Nickel

Most radionuclides of nickel form as activation products or the decay products of radioactive isotopes of other activation products, notably radionuclides of cobalt. Thus, cobalt and nickel are strongly connected in their radiochemistry. The direct discharge of radionuclides of nickel to the Great Lakes is infrequent, and a continuous discharge of radionuclides of nickel suggests possible reactor shielding operational problems.

Complicating the situation about nickel discharges to the Great Lakes is the INCO smelter at Sudbury, Ontario. The stack on this smelter is the largest discharger of nickel to the environment in North America. A gradient of nickel levels in water and terrestrial vegetation from Sudbury, Ontario, to the Great Lakes tracks these discharges. As an example, compare the nickel level in zooplankton from McFarlane Lake in Table 16 with other lakes in the Sudbury area. (Yan *et al.* 1989).

Stable nickel isotopes have relatively large thermal neutron absorption cross sections, making the activation of nickel in the environment by other radionuclides are of possible concern. The Task Force raises the question of whether the nonradioactive nickel in the smelter stack effluents that deposit in the Great Lakes following long-range transport are subject en route to nuclear activation. There are several possible mechanisms which the Task Force could postulate, but no information on any hypothesis is available nor is documentation.

The height of the smelter stack provides a high tropospheric discharge port for the gaseous effluents, and various atmospheric circulatory processes may move some of the gas and particulate matter to higher altitudes and exposure to cosmogenically derived neutrons. There may also be an intersection of stack plumes at some location between the smelter gaseous and particulate effluents and the similar gaseous and particulate effluents from a nuclear power plant. Another question relates to a possible activation processes by a within-lake (in situ) source of radionuclides. The Task Force does not have answers to the preceding questions, but raises them mainly because of the uniqueness of the sources of nickel to the Great Lakes.

The main radionuclide of nickel of interest is ^{63}Ni (half-life 92 years). A second and very much longer lived nuclide, ^{59}Ni (half-life 80,000 years), is occasionally of interest.

Nickel accumulates in aquatic organisms. A biological role for nickel as an essential nutrient may exist. Price and Morel (1990) cite nickel as a possible heavy metal cofactor for enzyme urease. DeFilippis and Pallaghy (1994) cite data on the freshwater red alga, *Cyanidium caldarium*, as showing some toxicity resistance to Cu/Ni combinations in laboratory cultures, suggesting either an exclusion mechanism for Ni uptake or the ability to immobilize the bioaccumulated materials.

TRANSURANIC ELEMENTS

The presence of transuranic elements as natural inorganic constituents of freshwater biota is rare. Cannon (1960) has studied the presence of transuranics, mainly uranium and thorium, in terrestrial plants. Cowgill has provided data on thorium in her study of the macrophytes of Linsely Pond. She also found thorium in the aphid (*R. nymphaeae*) which feeds on the leaves of the macrophytes and in the sediments, but none in the water column. Thus, the Task Force has not attempted any estimates of thorium uptake in biota from the water column, but has considered uptake estimates relative to sediments. This is actually a preferred approach since previously the Task Force indicated that transuranics move to sediments as their environmental repository. Uptake from the water column would mainly require ingestion to bring the transuranics internally into the organism as opposed to direct osmosis from the water column. It is possible that such ingestion would find the transuranics bound to particulate matter in the water ingested (Table 19).

TABLE 19
THORIUM IN AQUATIC ORGANISMS
(Adapted from Cowgill 1973*a*, 1973*b*)

Substrate	Thorium (ppm)		Bioaccumulation Factors	Enrichment Factors
Linsely Pond				
Soils	2.6			
Rocks	2.2			
Sediments				
Deep water	3.2			
N. odorata	0.88			
N. odorata (1971)				
Flowers	2.3	Flowers/sediments[2]	2.6	0.14[4]
Flower stalks	1.1	Flower stalks/sediments[2]	1.3	0.09[4]
Leaves	1.6	Leaves/sediments[2]	1.8	0.12[4]
Stems	1.5	Stems/sediments[2]	1.7	0.11[4]
N. odorata (1972)				
Flowers	1.7	Flowers/sediments[2]	1.7	0.11[4]
Flower stalks	0.6	Flower stalks/sediments[2]	0.68	0.04[4]
Leaves	1.7	Leaves/sediments[2]	1.9	0.13[4]
Stems	0.1	Stems/sediments[2]	0.17	0.01[4]
R. nymphaeae (aphid)	0.76	Insect/average plants[3]	0.46	0.03[4]
Outlet water	Not detected			
Crustal material	9.6[1]			

Notes: (1) References: Cowgill (1973), Taylor (1964). All data are dry weight.
(2) Sediments used in these calculations are those from which the macrophytes were harvested.
(3) Plants used in these calculations refer to leaves on which the aphid feeds.
(4) Reference element for the enrichment factor calculations is titanium. Thorium is not easily accumulated by either plants or insects. Under these circumstances, radiation effects of thorium on biota are reasoned to reflect internal irradiance, and not accumulation in tissues.

MODELING ANALYSIS OF FISH DATA

Dr. Jack Cornett of the Chalk River Laboratory of Atomic Energy Canada Limited supplied data for radionuclides in fishes. These data were in the form of the wet weight and therefore require some estimates on what might be the water content of material. To see how this affects calculated biological transfer factors, notably bioaccumulation and enrichment factors, a simple modeling analysis was undertaken. A range of the water contents of the substrates studied that were typical of biological materials were chosen, and the bioaccumulation factors on an *assumed dry weight basis* were calculated and compared.

Data are presented for 18 elements that have nuclides with half-lives of greater than 40 days. As can be seen, the difference in estimated bioaccumulation factors can vary over as much 1.5 orders of magnitude (Table 20). Several of the elements show unusual bioaccumulation possibilities in freshwater fishes, which have not been observed in other freshwater taxa, notably iodine, antimony, and zinc. Iodine numbers reflect the presence of thyroid hormones, a unique hormonal constituent in the vertebrates. Zinc is found as cofactor in many vertebrate hormonal systems, especially pancreatic materials since it is the activating element of insulin, and in a number of proteins in vertebrate systems which have a "zinc finger" in their structure (a group of four or five nitrogen atoms from amino acid groups bonded covalently to zinc as a means of stabilizing a particular conformation of protein with a finger or appendage design, or even a peninsula effect if the rest of the protein shape is compared to an island. Antimony is unexpected. Freshwater biota, mainly plants (algae) and invertebrates (zooplankton and insects) have shown no evidence of antimony, and the antimony data remain interesting and unexplained.

A number of elements in Table 20 show lower bioaccumulation potential than is exhibited in plants and invertebrates. Notable here are calcium, magnesium, potassium, molybdenum, and vanadium. Calcium, magnesium, and potassium reflect the physiological systems of fishes uniquely. Molybdenum and vanadium appear in vertebrate physiological systems but are more related to plants.

Several elements show greater bioaccumulation potential than would be expected, and this is also related to vertebrate physiology, but these elements have known bioaccumulation possibilities in all taxa, notably cesium, phosphorus, and rubidium. Cesium and phosphorus reflect that vertebrates have a calcium phosphate based skeletal structure, and the crystalline pattern of apatite minerals is adequate to accommodate the atomic and ionic radii of cesium, where it might not otherwise occur. The atomic and ionic radii of rubidium are smaller than comparable radii of cesium and can also fit into the mineral matrix of apatite. In addition, many rubidium compounds are water soluble and allowing the presence of this element in aqueous matrices.

TABLE 20
BIOACCUMULATION FACTORS FOR FISHES FROM LAKE HURON FOR VARIOUS ASSUMED WATER CONTENTS OF TISSUES

Element	**Fish Flesh[2] (mg/kg wet weight)**	**Water (ng/L)**	***R***	***R* assuming 50% water content in fish flesh**	***R* assuming 80% water content in fish flesh**
Ba	0.017 ± 0.014	14.2 ± 1.1	1.2	2.4	12
Ca	339 ± 416	25,640 ± 1942	13	26	130
Cr	0.053 ± 0.021	0.46 ± 0.08	114	228	1140
Cs	0.011 ± 0.006	0.001	10,800	21,600	216,000
Fe	4.4 ± 3.9	10.7 ± 0.7	415	830	4150
I	0.12 ± 0.04	0.96 ± 0.09	6076	12,152	60,760
K	4071 ± 485	670 ± 130	62	124	620
La	0.00041 ± 0.00024	0.005 ± 0.001	52	104	520
Mg	257 ± 37	5800 ± 875	44	88	440
Mn	0.11 ± 0.05	0.28 ± 0.17	412	824	4120
Mo	0.0048 ± 0.0021	0.41 ± 0.06	12	24	120
Na	407 ± 162	1.40 ± 0.23	146	292	1460
P	2374 ± 270	8 ± 5	29,600	59,200	296,000
Rb	6.3 ± 2.4	0.65 ± 0.11	9724	19,448	97,240
Sb	0.0059 ± 0.011	0.11 ± 0.11	53	106	530
Sr	0.223 ± 0.222	88 ± 9	2.54	5.08	26
V	0.10 ± 0.04	0.56 ± 0.27	155	310	1550
Zn	3.9 ± 0.9	0.72 ± 0.08	5510	10,200	102,000

Notes: (1) Data given in columns 1, 2, 3, and 4 are adapted from Table 3 of report WPTR No. 2719, *Reducing the Contribution of the Fish Pathway to DEIs* 1997 Annual Report (Contract Officer: Lorna Chant). Values are means ± standard deviation of the results. Since most biological samples can have large variability in elemental composition, reported standard deviations sometimes exceed the means. Such data indicate the data may be distributed "log normally" instead of "normally."

(2) While only fish flesh data are given herein, the referenced source of that data suggest that the bioaccumulation factors for Cs, K, Rb are the same for whole fish because these elements are uniformly distributed throughout the specimens.

(3) The data indicate a rare observation of Sb in tissue and water. The referenced source indicates that Sb was not detectable in whole-fish samples.

CONCLUDING REMARKS

This report has examined in detail bioaccumulation of elements in freshwater aquatic biota and selected species of terrestrial plants (mosses) and wildlife (caribou). The elements chosen are those that have sufficiently long-lived radioactive isotopes that they could reasonably be expected to cycle through the Great Lakes biotic communities and exert both the chemical effects associated with the element as well as any harm from α, β, and γ radiation that accompanies the isotope. The only α emitter for which any biotic data existed that met the tests of quality assurance and reportability of the Task Force was thorium. Most of the remaining isotopes covered are β emitters.

The report examined aquatic plants, zooplankton, and a terrestrial insect. Appropriate data for fishes were not available, although some model analyses of fish data supplied by Dr. Jack Cornett of the Chalk River Laboratory in Canada give a possible snapshot as to what bioaccumulation figures might be for fishes. In addition, selected fish data from Ontario Hydro's nuclear power stations on the Great Lakes were available for ^{40}K and ^{137}Cs. From these data, selected bioaccumulation factors were suggested.

The various data strongly suggest that bioaccumulation within freshwater food chains requires careful study. The behavior of many elements which have radioactive isotopes of half-lives greater than 40 days is often anomalous and unexpected. Many elements perform their physiological function as part of interconnected metabolic systems, and therefore, one must examine the behavior of several elements as a suite before assuming that a given bioaccumulation factor is appropriate. This is especially true when using data from marine species to make judgments about freshwater species.

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