Field measurements of the transfer factors for iodine and other trace elements

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ECOMatters Inc.

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ABSTRACT

Title: **Field measurements of the transfer factors for iodine and other trace elements**
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Abstract
Iodine-129 is one of the key radionuclides related to environmental assessments of nuclear fuel waste management because it has a very long half-life, is environmentally mobile, and iodine (I) is an essential element for animals. However, data are sparse because both ^{129}I and stable I are difficult to detect. In this study, a new method has been adopted that allows reliable measurement of stable I in almost all biological tissues. This method was used to survey transfer factors from media (water and soil) to a variety of Canadian plants and animals. In addition to iodine, up to 62 other elements were measured.

The plants and animals considered are relevant to the human food chain. The data include both traditional farm products, as well as a variety of wild fish and game. The food products related to modern agriculture included a comparison of garden versus field crops, as well as transfers to milk, eggs and meat. The survey of meat products included beef, pig and chicken and was augmented by single samples of rabbit, lamb, squab (domestic pigeon), turkey and domestic goose. The wild animal data is relevant to both a hunter lifestyle, as well as providing information on movement of iodine and trace elements through biota of general interest. The data included 9 species of wild fish as well as wild deer, geese and blueberries. Additional single samples of caribou, elk and moose extended the range of species. The resulting transfer factors were fish/water concentration ratios, meat/feed concentration ratios and feed-to-meat fractional transfer factors. Soil and sediment solid/liquid partition coefficients, a survey of iodine concentrations in 20 lakes and aquatic macrophyte/water concentration ratios were also completed.

In general, where comparisons to the literature were possible, there was good agreement. However, this study markedly increased the number of data for iodine over what was previously available, and coupled these with data from the same systems for many other elements of interest. There was strong evidence that meat transfer factors might be more consistently expressed as concentration ratios rather then the traditional fractional transfer factors.

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1. INTRODUCTION

Iodine-129 is a prominent radionuclide in the dose and risk estimates for disposal of high-level waste. This is because the radionuclide ^{129}I has a very long half-life and the element, I, is mobile and is essential for most biota, including humans. Despite its importance, transfer data for I (and hence ^{129}I) are scarce because the radioactive isotopes (^{125}I, ^{129}I and ^{131}I) are difficult to use and the stable element has been difficult to measure at environmentally relevant concentrations.

Although radioactive ^{129}I is the iodine (I) isotope of concern for long-term waste management, there is consensus that all isotopes of iodine will behave in the same manner in the environment, due to the small mass difference between the isotopes. Thus, measurement of stable iodine in both substrate and recipient media can be used to compute transfer factors to model the potential transfer of ^{129}I among those media (e.g., from soil to plant, plant to animal). Until recently, the most sensitive routine method to measure iodine in environmental samples was by instrumental neutron activation analysis (INAA), giving a detection limit of about 1 mg kg^{-1} (1 ppm) in solids. This is not sufficiently sensitive to measure iodine in environmental samples except in materials such as seaweed or in plants grown near the ocean. Accelerator mass spectroscopy (AMS) was more sensitive, but expensive with limited availability. Normal thermal ashing of samples, often used to improve detection limits, cannot be used for iodine because of its volatility. Thus, measurement of transfer factors was generally limited to experiments where radioactive isotopes could be used, or where stable iodine levels were elevated above normal. These methods are both artificial and expensive.

Several recent advances have been made in the measurement of iodine. Inductively coupled plasma mass spectroscopy (ICP-MS) has a good detection level for I in solution, of about 1 mg L^{-1} (1 ppm). Various methods have been used to dissolve or extract iodine from solids so that ICP-MS or other liquid-based methods can be used for measurement. These include dry ashing with capture of the off-gas, dry ashing in Parr bomb (sealed container), dry ashing with alkaline additives, and alkaline extraction such as with tetramethylammonium hydroxide (Fecher et al. 1998, DIN EN ISO 17294-1 2005) – “the German Food Standard method – or GFS”. Detection levels as low as 2 µg kg^{-1} (2 ppb) in solids have been reported. In the first phase of this project, the GFS method was proven to provide good detection for iodine at these levels at a reasonable cost (Appendix A). These detection limits will allow measurement of iodine transfer factors in most plant and animal tissues.

The intent of this project was to utilize the proven GFS analytical method to measure key transfer factors and to obtain other ancillary media parameters, such as soil Kd, in three food chain systems – aquatic, agricultural and terrestrial hunter/gatherer. This work will contribute to a comprehensive international database of transfer factors for iodine, as well as provide Canadian specific data.

1.1 OBJECTIVES

The primary objective was to obtain biosphere transfer factors from natural and farm settings in Canada, with emphasis on iodine but to include as many other trace elements as practical. The fundamental supposition is that transfer factors measured with stable elements are good surrogates for transfer factors of long-lived radionuclides that may be emitted to the environment at low rates over very long time periods. The transfer factors include wild game, farm animals and cultivated plants, and supporting data such as soil/liquid partition coefficients for soils and sediments. An equally important outcome is the reporting of background concentrations of trace elements important to nuclear waste management, especially I and Cl.

2. METHODS

2.1 GENERAL STRUCTURE OF THE STUDY

The sampling began in 2006 with proof of concept, testing the ability to reliably quantify iodine in various environmental media. In 2007 and continuing into 2008, the emphasis was on sampling of wild biota important to a hunter lifestyle in a Canadian Shield or southern Canada area, so this included wild deer, geese, fish and blueberries, with opportunistic sampling of elk, caribou and moose. Gut contents of terrestrial animals were used as the surrogates for ingested feed – the substrate for transfers. Fish and other aquatic biota, nominally 5 species per site, were sampled from a series of water bodies across Ontario and Manitoba, and were supplemented by samples from a lake in the Shield in the North West Territories. In 2008 and 2009, the emphasis was on typical Canadian farm animals including dairy, beef, swine, and layer and meat poultry, with some sampling of other species directed to obtaining samples from a range of body sizes, and sampling from a coastal region (Nova Scotia) to contrast with sampling mid-continent (Manitoba). Ingested feed sampled as reported by the farmers was sampled as the substrate for transfers. Additionally in 2008, crop sampling was conducted with the objective to obtain both field and multiple garden crops from the same soil types, in order to be able to compare transfers to different crops in the same settings. In 2009, samples were also obtained of honey, pollen, bees and adjacent flowering crops. Ancillary data included moisture contents and animal feed intakes rates (where possible) in order to present transfer factors in the units required by assessment models.

In addition to sampling to measure transfer factors, concentrations of I and other trace elements were measured in 20 other water bodies. These samples were taken to increase the availability of background concentration data, which are sparse especially for I.

2.2 STATISTICAL DESIGN

The numbers of samples were limited by the costs of iodine analysis, and although this limited the statistical power, use was made of good global information on the expected error variance. The experimental design for samples of fish and other aquatic biota was randomized partial factorial with location and species the categorical factors. Not all species were represented in all water bodies. The experimental design for terrestrial animal samples was nominally a completely randomized design with species as the categorical factor. Effects of sample source and animal size were evaluated based on means comparisons and correlation or regression analysis. Effects of date of sampling were considered part of the experimental error. The experimental design for soil-to-plant transfers was a completely randomized design with plant type (grain, leafy, fruit and root) as the main factor. Location effects were considered part of the experimental error, as was species within the various plant types.

The default assumption for the data frequency distribution of ratio data was lognormal, consistent with previous experimental evidence and with the Central Limit Theorem. For ratio data and any other data deemed to be lognormally distributed, all statistical tests including correlations and regressions were of log-transformed data, and geometric means (GMs) and geometric standard deviations (GSDs) are reported.

2.3 GENERAL DEFINITION AND COMPUTATION OF TRANSFER FACTORS

Two types of transfer factors are computed. The simplest are concentration ratios. The aquatic-biota/water concentration ratio (B) is the concentration in the biota, usually the edible portion, on a fresh weight (FW) basis divided by the concentration in the substrate water. The resulting units for B used in this report are L kg^{-1}(FW), although with the assumption that the density of water is 1 L kg^{-1}, B is sometimes reported in the literature as unitless. The plant/soil concentration ratio (C_r) is computed as the concentration in the dry plant, for a specified plant part that is usually the edible portion, divided by the concentration in the dry substrate soil to 30-cm depth. Because the units of concentration are the same, C_r is unitless. The solid/liquid partition coefficient (Kd) for sediment or soil is the concentration on the dry solids divided by the concentration in the specified corresponding liquid, ideally the pore water but in the case of sediment it is sometimes the overlying water. The resulting units for Kd used in this report are L kg^{-1}, although m^3 kg^{-1} would be the SI units.

Although not commonly used at present, animal/feed concentration ratios may be used in future assessment models for certain elements, and are usable when the net level of contamination of the complete animal feed is known. These ratios are computed as the concentration in the biota, usually the edible meat, egg or milk, on a fresh weight (FW) basis divided by the concentration in the complete feed or stomach content on a dry weight (DW) basis. For milk, the resulting units are kg(DW) L^{-1} and for meat are kg(DW) kg^{-1}(FW).

The other type of transfer factor, and the one most commonly used for terrestrial animal transfers, are computed as fractional transfer per day (F), classically computed as the concentration in the meat (F^{meat}), milk (F^{milk}) or egg (F^{egg}) product divided by the daily intake of the specified element or radionuclide. The units are d kg^{-1}(FW) for meat and egg and d L^{-1} for milk. However, when the daily intake of the element can be represented by the concentration in the whole feed divided by the daily intake of whole feed, F is equivalent to the (meat, milk or egg)/feed concentration ratio, as described above, divided by the daily feed intake. Because feed intake is related to body size, values of F are expected to vary with body size, both within and between species.

Daily intake is directly quantifiable for some farm animals, but not for wild animals or farm animals allowed to feed freely. For these, it is necessary to estimate feed intake from body size, and allometric functions to do this are reported in the literature (Macdonald 1996).

Because of the uncertainties associated with sampling feed as consumed and uncertainties in quantifying daily feed intake, manure from farm animals was also sampled. These data are not directly relevant to computing transfer factors, but are useful to identify or explain anomalies. For example, if in one animal some component of the diet such as a livestock mineral supplement was not sampled, or its portion in the total diet was not accurately accounted, then this may be revealed by different manure/feed concentration ratios for this animal compared to other animals of the same species.

In addition to samples of diet and food product, samples of the animal drinking water were collected, largely to assure that water is a not a significant source of I or other trace elements.

2.4 SITE SELECTION AND SAMPLE COLLECTION

2.4.1 Aquatic Biota and Survey of Water Concentrations

For aquatic biota, the sampling strategy was to obtain 5 species from each water body, and this was to include one macrophyte and 4 fish species. The fish were to represent a range of body sizes and ecological niches, and to have some common species among the sampling sites. In addition to the lakes sampled for fish, 20 other Canadian Shield lakes were sampled to measure background I concentrations.

For sampling convenience, most of the Canadian Shield water bodies sampled were in Northern Ontario, in areas accessible from the main TransCanada highway. To balance our survey spatially, we divided the area into five (roughly equal by longitudinal distance) sub-sampling zones (Table 1 and Figure 1). Each zone was reasonably distinct, with key features shown in Table 1. Zone 1 also contains the Experimental Lakes Area (ELA), and staff of the Department of Fisheries and Oceans, Winnipeg supplied both archived and present-day samples from ELA.

Table 1. Geographic features of each of the five sampling zones.

Zone 1	Large rocky granite outcrops; jack pine dominant tree species; many small and large oligotrophic lakes
Zone 2	Flat to lowland; sandy and clay soils; mixed coniferous forest; fewer, larger, shallower lakes
Zone 3	Significant topographical relief; rivers draining the Lake Superior watershed dominate; jack pine dominant tree species
Zone 4	Flatland to lowland interspersed with periodic granite outcrops
Zone 5	Transition to agricultural land; interspersed with granite outcrops; mixed forest dominant

Figure 2-1. Outline map of the Province of Ontario, overlain with crude polygons illustrating each of five sampling zones.

In addition to the Canadian Shield samples taken in Ontario, further biota and water samples were taken in Sylvia Lake (essentially a hydroelectric impoundment) along the Winnipeg River in Manitoba, near-shore in Lake Huron near Tiverton Ontario, and Mackay Lake in the North West Territories (NWT, about 100 km north-east of Great Slave Lake).

Sampling in Ontario, with the exception of ELA and Lake Huron, in Manitoba and in the NWT was done by staff of ECOMatters. Ontario Ministry of Natural Resources (OMNR) scientific collection permits were obtained to allow sampling with nets in specified water bodies in each zone in Ontario. From the list of permitted lakes, we could choose in the field which water body to sample, according to accessibility and specific fish habitat characteristics more readily identified on-site than from a remote location. Similarly, a permit was required and obtained from the Aurora Research Institute in Inuvik to remove water samples from Mackay Lake. Angling permits were obtained for hook-and-line fish sampling in Manitoba and NWT. Permits for sampling in the ELA were the responsibility of staff from the Department of Fisheries and Oceans in Winnipeg, who did the sampling. Permits for sampling in Lake Huron were the responsibility of Bruce Power and their contractors, who also did the sampling.

For water sampling, ideally more than one position was sampled within the same water body to produce a composite sample. Expected water depths were estimated based on lake basin geomorphology, local knowledge and a varying quality of maps. Actual water depths were determined using a standard fish-finder in combination with manual soundings taken during sampling. Water samples were collected using the following materials and methods:

1. Two plastic 4-L collection / mixing pails were rinsed with local water prior to sampling.

2. A 2-L Kemmerer sampling device held on a 25-m-long graduated line was rinsed with local water prior to sampling.

3. We estimated the depths at which thermal stratification might be expected.

4. Surface (epilimnion) water temperature was taken using a digital thermometer (Control Company - Traceable CE -50° to 150°C). This value was used to validate / refute our depth estimates for both the metalimnion (depth layer in the lake with mid-range temperature) and hypolimnion (lowest depth layer in the lake with lowest summer temperature), based on the difference in temperature compared to epilimnion (surface) readings.

5. On-site depth estimates of the thermocline were made and a preliminary sample was taken at a depth we expected would represent the metalimnion. This water was stored in a collection pail. A second preliminary sample was taken at a depth we expected would represent the hypolimnion. This water was stored in a separate collection pail. Once we had established depths associated with clear thermal stratification, we proceeded to collect final water samples.

6. Final water samples were collected from each depth using the methods just described. During sampling, water was stored out of direct sunlight and temperatures monitored to ensure sample stability. We discarded samples where temperatures changed by more than 2°C while sampling, and then these were re-sampled. To produce composite samples a second (and sometimes third) sample was taken at the same depths from proximate (usually within 1500 m) sampling points. Samples were pooled by depth (i.e. metalimnion samples or hypolimnion samples) and prepared for field processing.

7. Final pooled samples were shaken to mix the water thoroughly. Water was extracted from each composited sample using a separate syringe for each raw sample. Two 30-mL glass scintillation vials were filled approximately 2/3 full after passing the raw sample through a 0.45-µm Whatman filter. We stabilised one of these samples for macro- and trace-elements analysis using 1-5 drops of 5% HCl and ensured sufficient acidification using pH test strips ($pH \leq 2$). The other sample was for I analysis and was not acidified. The filtering and preservation process was repeated on both metalimnion and hypolimnion composite samples.

8. Water samples were labelled and stored out of direct sunlight while in the boat and stored in the deep freeze upon return to shore.

To represent macrophytes, we selected *Stuckenia* (*Potamogeton*) as a meaningful representative plant genus that is abundant, ubiquitous to the boreal biome and relatively easy to sample. Rooted macrophytes in general, and *Stuckenia* in particular, have a direct linkage to the sediments in which they grow. They also possess considerable leaf surface area exposed to the water column. *Stuckenia* was sampled only in lakes in Northern Ontario in which fish were sampled, and were collected near the fishing locations. To sample, we grasped floating stalks by hand and with gentle pressure usually were able to extract several individual plants from the sediment. Labelled macrophyte samples were kept out of direct sunlight and stored in the deep freeze upon return to shore.

A single water body within each sampling zone in Ontario was selected for fish sampling. We chose larger lakes wherever possible, with the expectation that they would have a richer ichthyofauna to sample and therefore provide a better chance of sampling all of our target species. Prior to fish sampling, we selected 5 target species or genera that were 1) expected to occur across the sampling area; 2) occurred at higher levels of the trophic order; 3) formed part of the human food chain or act as sport fish that are sometimes consumed by humans; and 4) showed different feeding patterns to encapsulate as many pathways as possible within the aquatic environment. These target species / genera are listed below (Table 2). Fish were captured using both passive and active gear (Table 2). We used gill nets (both clear monofilament and cotton mesh) as the most effective passive gear type for capturing larger fishes (Arthaud, 1992). Short set times (0.5 - 4 hr) were used to minimise by-catch mortality. We also used a 4-mm mesh size 5- x 5-m beach seine to capture both juvenile large-bodied fish and small bodied fish species. Generally we could capture 3 of our 5 target species with comparative ease; the further species required considerable additional effort.

Table 2. Dimensions of passive and active fishing gear used to capture fish samples, July - August 2007. Common and scientific names of target species / genera based on net mesh size and water depth (based on hanging depth) also given.

Mesh size (cm)	Hanging depth (cm)	Mesh Type	Length (m)	Target Species /Genera
5.1	240	green (cotton)	45	Smallmouth bass (*Micropterus dolomieu*) Yellow perch (*Perca flavescens*)
8.9	180	clear mono-filament (braided)	90	Sucker spp (*Catostomus / Moxostoma*) Walleye (*Sander vitreus*) Northern pike (*Esox lucius*) Lake whitefish (*Coregonus clupeaformis*)
8.9	360	clear mono-filament (braided)	70	Walleye Northern pike Lake Trout (*Salvelinus namaycush*)
0.4	90	white cotton	5	Emerald shiner - (*Notropis atherinoides*) Juvenile of large-bodied species

Gill nets were always set perpendicular to shore and were set from the shoreline itself out for the full length of the net. In all cases, nets were marked with orange buoys, marked with OMNR permit information. Setting nets in this fashion required considerably more effort than using a pelagic free-floating method. Wherever we used multiple nets and where setting was complicated, we were sometimes obliged to return to earlier set net(s) to check for fish, tangles or problems before completing setting of subsequent nets. On the other hand, when setting was relatively quick, we were able to collect water samples during the time nets were working. Periodically, we tried free floating pelagic netting, but short set times meant that we achieved nil fishing success using this method. Once nets were set, we recorded the time (to monitor set

times) and position using a Garmin 12XL GPS hand held unit. In general, we remained in close proximity to nets to prevent human interactions, either deliberate or accidental. Captured fish were retained in a live well, identified to species, and measured for total length. We retained only sufficient numbers of individuals per species to produce 200 g wet weight edible tissue samples. Fish were filleted on shore and excess fish tissue was disposed of according to the directions in our OMNR permits. Labelled samples were stored out of direct sunlight and stored immediately in the deep freeze.

Aquatic samples were obtained primarily by use of a small boat, but some samples were obtained from shore or by wading. Since sampling was conducted in summer across distances greater than 2500 km, sample preservation was of special concern. To stabilise samples as much as possible, we transported a small (0.5 m^3) deep freeze in our research vehicle. Cold freezer temperatures were maintained using ice packs during the day and the freezer was plugged in to AC electrical power each night. The thermal mass of the ice (initially, and then samples as they replaced ice) kept the samples from spoiling.

Sampling in Mackay Lake NWT was as above, except that fish were obtained by angling. No macrophytes were sampled.

From the ELA, we obtained archived frozen samples of whole Northern Pike and Lake Trout from a single lake. These fish were part of an earlier independent project, and were taken in 1999, 2001, 2003, 2005 and 2007. In 2008, additional fish samples and water samples from the same lake were obtained specifically for our use. All fish were filleted in the ECOMatters lab to collect the usual edible muscle tissue and a composite of pike heart and liver tissue from years 1999 to 2007 (a composite was required to obtain enough tissue). Water was filtered to pass 0.45 µm and stored in glass containers as above, with one portion acidified and the other stored frozen. No macrophytes or sediments were sampled.

Sampling in Lake Huron was done in conjunction with the monitoring program of Bruce Power, with samples drawn from 2 regions in the lake, both considered background relative to the nuclear facility. No macrophytes or sediments were sampled. The samples were shipped by courier to ECOMatters. Filtered water samples were not frozen but instead were shipped immediately to the analytical laboratory. These were in glass bottles, sealed with no headspace. Fish fillets were shipped frozen to ECOMatters.

2.4.2 Sediment

Sediments were collected using either of two methods. Sampling the hypolimnion of lakes where depth was <25 m usually resulted in the capture of soft sediments in the Kemmerer sampler. These sediments were either poured off directly into collection bags and used as a final, discrete sample, or alternatively, poured into collection pails to form composite samples, depending on the quantity of sediment captured. Clearly, profundal sediment samples of deeper lakes (e.g. Quirke Lake) were not possible using this method, and could not be sampled. In other cases, sediments were not soft and could not be obtained using the Kemmerer sampler. Often, in these latter cases, we collected littoral (shallow-water) sediment samples using a graduated telescoping pole to which was attached a scoop. Labelled sediment samples were kept out of direct sunlight and stored frozen.

2.4.3 Wild Geese

Five Canada Geese (*Branta canadensis*) were sampled, from one hunter who harvested all samples on one day from Oak Hammock Marsh in Manitoba (October, 2007). The hunter provided us with the viscera and a thigh from each bird for analysis. It is not possible to know if these geese originated from the same or different summer feeding habitats, but it is likely that their recent feeding was in similar habitats. Iodine has a relatively short biological half-life, so that tissue concentrations probably reflect recent feeding, in the order of weeks.

2.4.4 Wild Ungulates (Deer, Moose, Elk, Caribou)

Ungulates were sampled by 6 hunters, half of whom supplied multiple samples. Each hunter was provided with identical written instructions and sampling kits. Briefly, hunters were instructed to provide both the contents of the first stomach and approximately 200 g of belly meat from legally harvested ungulates. They were provided laboratory gloves to handle the samples. Ungulate samples were primarily white-tail deer (*Odocoileus virginianus*) harvested from the Whiteshell Park area of Manitoba. However, elk (*Cervus elaphus*) and moose (*Alces americanus*) samples were provided from the Duck Mountain area of Manitoba. Meat of a caribou (*Rangifer tarandus*) from the Mackay Lake NWT area was provided, but stomach contents were not available. In this case, possible browse vegetation and caribou scat were sampled using a random walk sampling pattern in an area frequented by caribou. Samples were delivered fresh or frozen to ECOMatters for further processing.

2.4.5 Dairy

Sampling of dairy farms was divided between mid-continent (Manitoba) and coastal (Nova Scotia) in order to ensure some concentration range in the samples. Manitoba is an inherently low-iodine region, largely because it is far from the oceanic source of atmospheric iodine. Nova Scotia is a high-iodine region because of the proximity to the ocean. Previous sampling in 2006 indicated up to 100-fold higher plant concentrations in Nova Scotia compared to Manitoba. It is recognized that for dairy, commercial feed is commonly used and iodine and other mineral supplements will be included. However, dairy in particular will be fed local forages. It is also recognized that iodine teat dips are nearly universal in dairy (to prevent mastitis), and so special attention was made to sample cows without teat dipping prior to milking.

The primary criteria for selection of farms to sample, apart from Province, were cooperative farmers within ~200 km of our base who were typical commercial producers. Provincial agricultural advisors, University staff, commercial agricultural consultants and word of mouth were used to find cooperative farmers. When enquiring about farms in Nova Scotia, an offer was made to allow us to sample the dairy herd on the campus of the Nova Scotia Agricultural College (NSAC, professors are adjunct to Dalhousie University). These cows are fed and housed in the same way as commercial animals. The advantage of utilising this source is that there was very detailed record keeping and there were skilled technicians available who understood sampling protocols and who were well suited to sample in our absence.

In general, dairy cattle (*Bos primigenius*) are milked twice each day, which means they are in barns and are handled during this time. Most milking cows stay in barns all day. Typically, one stock of feed is blended and given to all the cattle at milking time. This blended feed contains farm-grown forages, farm-grown or locally grown grains and commercial feed supplements. Teat dips are used prior to milking and all but one farm sampled used iodide compounds for this.

Each dairy farm was sampled twice, several months apart. The general sampling procedure was for the farmer to identify a cow that he would milk separately, usually into a ~40 L can designed for milking a single cow. In many cases this was an apparently random selection, in some cases it was a cow whose milk was kept separate for a reason, such as she had recently freshened (given birth). The teat was wiped but not dipped, the milk collected in the farmer's can and then transferred to a clean 1-L glass bottle. Often the remaining milk was discarded. The feed was sampled either in the trough in front of the cow or at the feed mixing station, collecting about 2 L of feed into a plastic bag, using gloved hands. The animal drinking water was sampled from an outlet in the barn as close to the animal waterers as possible, again into a clean 1-L glass bottle. Manure was often sampled directly from the same cow as milked: she would often defecate immediately after being disturbed and the manure could be collected as it fell with adept use of a plastic bag. In a few cases, manure had to be collected from the barn floor, and in one case the manure sample was extracted from within the body cavity. In all cases, gloves were worn during manure sampling. Cattle manure is semi-solid, care was take to avoid loss of liquid separates.

The milk and water samples were kept in the dark and frozen as soon as possible, typically in <12 hours. Manure samples that could not be dried immediately were also frozen for shipping and storage. There was a problem with breakage of glass jars when they were frozen, depending on speed of freezing, fullness of the bottles and other factors that seemed random. To avoid the risk of lost samples, several of the liquids sampled in Nova Scotia were put instead into washed stainless steel jars. In order to quantify the impact of this on analysis, several samples were split with one portion stored in glass and the other in stainless steel.

There were a few deviations from this general procedure. In some cases, the various components of the feed were sampled separately, and the mixing ratios (by weight) were obtained from the farmer. For these samples, we created a mixed feed in the laboratory using these mixing ratios. Similarly, in one case, cows had access to pasture as an exercise area, so a sample of the grasses present were collected. It was not possible to quantify what contribution grazing had to the total diet, but the farmer was convinced it was minor (the pasture was low-grade and the cows were otherwise well fed).

Notes were kept of features of the farm that could affect results. These included copies of some of the commercial feed analysis filed by the farmer.

2.4.6 Eggs

Sampling of layer farms was also divided between mid-continent (Manitoba) and coastal (Nova Scotia). It is recognised that some of the grain in poultry diets in Nova Scotia will have been shipped from central or western Canada, and so may not be as high in iodine as would locally grown grain. As with dairy, all layer farms were sampled twice, several months apart.

In general, layer hens (*Gallus domesticus*) are in cages with several birds per cage. There is a feed trough and waterer, the eggs fall through the cage and roll to the front, and the manure falls through the cage to a conveyor system. All birds in a barn are fed the same feed, and in most cases this is entirely commercially blended feed which will include mineral supplements. There are typically more than 10,000 birds per barn. Biosecurity is an issue for some farmers and for NSAC, so on these farms we were not allowed to enter the barns and the farmer did the sampling.

One to two dozen eggs were collected from the barn, usually unwashed. The eggs were refrigerated for storage and shipping, and then were washed, cracked, the yolk and white separated, and both parts weighed. The separates were then either immediately dried in a food dehydrator (<35°C) or frozen to await processing. Water and feed were collected as for dairy. Manure was either collected off the conveyor system under the cages or from the pile at the end of the barn. Layer manure is solid, so fluid loss was not a problem.

There were differences in age of birds, and in some cases where we sampled a farm the second time, this was of a different flock in the same barns (layer flocks are changed after about a year). Notes included in some cases the farmer's estimate of daily feed consumption per bird.

2.4.7 Livestock Meat Products

Most samples were of broiler chicken (*G. domesticus*), beef (*Bos primigenius*) and pork (*Sus scrofa*, also classified by some as *Sus domesticus*). Samples of lamb (*Ovis aries*) and rabbit (*Oryctolagus cuniculus*) were included to encompass a broader size range. A range of bird sizes was also obtained, including squab (domestic pigeon, *Columba livia*), Cornish hen (*G. domesticus*), goose (*Anser anser*) and turkey (*Melleagris gallopavo*). The challenge was to find farmers who slaughtered their own animals and had meat available. In some cases, the meat cuts were kept by the farmer for their own consumption or were to be sold 'at the farm gate'. In some cases, the animals were specifically killed for sampling, or were culls or dead stock.

Broiler chickens are generally raised in barns on straw litter and fed commercially prepared feeds. We did include one free-range operation, where chickens foraged for some of their feed. Most were from Manitoba, with one sample from Ontario. Feed was usually sampled from the stock source on the farm. Water was sampled from an outlet that had the same water source as for the waterers. Manure was usually picked by (gloved) hand, with care to avoid straw bedding. In one case, manure was extracted from the colons of several slaughtered birds. The meat was provided either as whole dressed birds or whole birds. Breast meat (white) and thigh meat (dark) were retained as two separate samples.

Beef usually begin life on pasture but as they near finishing they are usually housed in feedlots. Grazing or home-grown forages and grains constitute most of the diet. Salt licks provide mineral supplementation. Most beef samples were from Manitoba, with one from Ontario. Water was sampled from the waterers (outdoors) or from an outlet with the same water source. For beef in feedlots, which was most common, the feed sampling was typically of separate samples of stored forages, farm-grown grains, and commercial mineral supplement. Usually it was well known which forage and grain the animals had consumed in their last month, and the farmer could estimate the blend of feed materials and the typically daily consumption. For beef on pasture, a sample of the pasture grasses was collected. Some samples of salt licks were

obtained. Manure was collected where possible, including pats on pasture or in feed lots, and in one case from the colon of the slaughtered animal. The meat samples were prepared cuts, typically steak, ground beef or a roast. In one case, the animals were culled because of tuberculosis, and the inspecting veterinarian collected a sample of rump meat. One pair of samples was from the research herd at the Agriculture and Agri-Food Canada (AAFC) Research Station in Brandon Manitoba. The AAFC meat samples were delivered frozen to ECOMatters and the manure and feed samples were previously dried and ground at the research station.

Pigs are generally raised in barns with manure handled as liquid slurry. Feed may contain some home-grown feeds, but always includes commercial supplements. Most pig samples were from Manitoba, with one from Ontario. The manure was sampled as semi-solid from the pen floor or as liquid from the slurry collection pit. Meat was provided as prepared cuts or as a section of undressed animal, and so there was no uniform cut of meat retained as a sample.

The lamb sample and associated feed, manure and water was collected in the same way as beef. The farm was in Ontario. The lambs were fed dried forages with a mineral supplement. The rabbit meat and feed samples were from caged animals fed commercially dried alfalfa pellets, also from Ontario.

The Cornish hen samples were collected in the same way as broiler chickens, differentiating breast (white) and thigh (dark) meat, with samples from farms in both Manitoba and Ontario. The pigeon was from a farm in Ontario: two birds were sacrificed and all the meat that could be removed from both birds constituted a single sample. The domestic goose was from the same farm as the pigeon, and used the same feed as the pigeons. The two turkey samples were from NSAC in Nova Scotia and were from two birds in the same flock.

2.4.8 Wild Blueberries

Common blueberry (*Vaccinium myrtilloides*) was selected as a representative berry species that would be used in a hunter/gatherer community as well as a modern community. It is ubiquitous across much of Northern Canada. Two berry sites were picked per sampling zone, and these sites were chosen opportunistically based on presence of fruit. The berries were hand-picked using latex laboratory gloves. Approximately 200 g of blueberries were picked per site and placed in plastic bags. A corresponding soil sample from within the berry patch was collected either by scraping surficial soils where soils were sparse or by taking 10- to15-cm depth core samples using a standard agricultural soil sampler. Labelled berry and soil samples were kept out of direct sunlight and stored frozen. Samples of blueberry plants (comprising woody stems and leaves) were also collected, pressed and partially dried.

2.4.9 Crops

For the soil-to-plant transfers, the basic strategy was to sample several different plant types within each setting to give some information about how many types of plants should be invoked in an assessment model. In particular, it was considered important to obtain samples of field crops (cereal grains as the key example) paired with samples of garden plants. Essentially, this meant sampling farm gardens.

Most samples were collected by ECOMatters staff in Manitoba, but several farms were sampled in the area of Thunder Bay because in 2007 this area was found to be especially low in iodine. The sampling in Thunder Bay was by staff of the Thunder Bay Agricultural Research Station. The site selection criteria were largely set by the co-occurrence of garden and field crops in close proximity.

An additional criterion for sampling was to obtain some samples where air-exposed and occluded tissues could be sampled from the same plant. This may be especially relevant for I, some portion of which is deposited from the atmosphere. For example, cabbage allowed us to sample outer leaves that were exposed to the atmosphere and inner leaves that were never exposed. Similarly, corn husks were exposed but corn grain not.

Plant samples were collected by hand in a manner consistent with normal harvest, in that only the edible portions were sampled. Plant samples were refrigerated and shipped fresh (not dried or frozen) to ECOMatters. If processing could be done immediately, samples were stored under refrigeration. In the lab, samples that may have contacted soil were washed with deionised water. Cereal grains and interior tissues such as sweet corn and interior cabbage leaves were not washed.

Soil samples were collected as a composite of the top 30 cm of the soil in the area encompassed by the plant sampling. In some cases, separate soil samples for the field crop and garden crop areas were collected, despite there being no other obvious differences between these soils. There may have been differences in past and present fertilization and manuring practices. Soil samples were shipped to ECOMatters at field moisture content, not frozen.

2.4.10 Honey

The sampling of honey was largely considered a scoping activity, to determine if honey is a sufficiently unique pathway that further experimental work is required. In a safety assessment case, especially for a contemporary facility, one attribute about honey as a contaminant pathway is that the bees may be a specific vector that can conceivably move radionuclides from a protected area directly to a food product outside the exclusion zone. COG (2008) invokes a honey pathway. An example would be bees foraging in wildflowers in an exclusion zone immediately adjacent to a nuclear facility and then transporting contaminants in pollen and nectar to hives outside the exclusion zone, where honey is harvested for consumption. Another rationale for sampling the bees themselves is that they represent a plausible valued ecosystem component for assessment, either as honey producers or as representatives of an insect group.

Samples were collected near Pinawa Manitoba from bee yards in each of two fields, one was canola (*Brassica napus*) and the other was alfalfa (*Medicago sativa*). Although other flowering plants may have been present nearby, it was assumed that the large acreage of flowering crop provided most of the nectar and pollen collected by the bees (*Apis mellifera*).

Firstly, pollen was collected by inserting a commercial pollen collector in the entrance slot to one hive. The pollen collector was installed and left open, without the collection insert, for several days to ensure the bees accepted its presence. The collection insert was then put into position. The insert was essentially a perforated plate with triangular holes just large enough

for the bees to pass. In doing so, the bees lose the clumps of pollen they have gathered on their rear legs, and these clumps and fall into a tray. The amount of pollen collected depended on many factors, and this determined the sampling time for the honey.

Once 5-10 g of pollen had been gathered, the honey was sampled by disassembling the hive, removing a frame and scraping out a portion of honey plus comb. At the same time, a small jar was use to capture as many bees as possible, and a sample of the flowering crop plants was gathered, collecting the upper 30-cm portion of the plants with open blooms.

In the lab, the honey was separated from the comb by passing it through a screen, and the honey was retained for analysis. The bees were killed by freezing. The plants were weighed, dehydrated at <35°C, weighted again and ground in a knife mill for analysis. The pollen samples had barely sufficient mass, and so were handled with care to avoid loss. They were ground by hand in a mortar and pestle. Duplicates were prepared of one honey sample and the corresponding pollen and plant samples.

2.5 SAMPLE HANDLING

Wherever possible, laboratory gloves were worn to reduce contamination of samples by skin contact. Liquid samples, including milk and slurry manure, were collected in glass or stainless steel jars[1] that had been washed in a dishwasher and then rinsed with de-ionized water. Washing included washing of the jar lids. Water samples were filtered to pass 0.45 µm, and if not done in the field this was done in the laboratory. There were two protocols for liquid sample preservation. Samples for I and Cl analysis were frozen immediately and kept frozen except for one thawing to allow sub-sampling. Samples for trace element analysis of other than I and Cl were thawed, sub-sampled as needed, acidified with HCl or HNO_3 and then refrozen. Slurry manure samples were handled in the same way, but not acidified. Care was taken to ensure the samples remained frozen during shipment to the analytical laboratory.

Solid samples, including semi-solid manure, were collected in new, food grade, resealable plastic bags.

Samples of egg whites and yolks, meat, manure and bees were stored frozen until processed. Plant and soil samples were stored at room temperature until processed. Processing of egg samples was as described above, with numbers and weights of yolks and whites recorded separately. Meat samples, including fish, were thawed for processing, red meats were trimmed of fat, and all meats were cut into small very thin pieces. All meat, plant, egg, feed, bee, sediment and soil samples were weighed fresh, spread thinly on no-stick (teflon coated) aluminum foil and dried in a plastic domestic food dehydrator with forced air at <35°C. Drying usually continued for several days or until there was no further weight loss. Once dried, the samples were weighed again to compute the original moisture content. Unless already finely divided, dried samples were then ground in a small knife mill (a domestic coffee mill) to render a coarse powder that could be representatively sub-sampled in tenth-of-gram amounts.

Care was taken to use specifically designated dehydrator trays and knife mills for any feeds known to have mineral supplements that might contaminate other samples.

[1] Paired samples in glass and stainless steel were also collected to determine if the container affected the results.

Solid and semi-solid manure samples were spread on aluminum trays and dried outdoors, sheltered from rain, for up to one month. The samples were sent to the laboratory for them to grind and analyse. The laboratory was asked to return the unused sample for archive purposes.

Honey, once separated from the comb wax, was sent in glass bottles for analysis. These were stored and shipped at room temperature with no acidification or other processing.

During grinding, operators wore gloves, grinders were thoroughly rinsed with deionised water between samples, and new aluminum foil was used for each sample. Grinding was done with the grinder inside a plastic bag to minimize the potential for dust from one sample contaminating another (except for the manure samples that were ground by the analytical laboratory). Dried samples were stored in new, food grade resealable plastic bags.

2.5.1 Transfer Factors for Wild Geese and Ungulates

Because geese ingest grit (typically coarse sand) to aid in comminution and digestion, their stomach contents contained grit and this would bias the concentrations relative to the plant material actually consumed. It was impossible to remove the grit because some could have been too finely divided to be distinguishable from the other stomach contents. In effect, the grit contributes elemental content and mass. For iodine, we anticipate negligible contribution of iodine from the grit, and so correction for the effect of the mass of the grit yields an accurate transfer factor for plant browse to muscle. For other elements, notably cations, the grit will have contributed an unknown amount of the element, and so the transfer factor is for total ingested material to muscle. The correction applied to the data here (where indicated) was simply a mass correction based on loss-on-ignition analysis of stomach contents (88% of the stomach content was ash, and assuming plant material is no more than 15% ash, 73% of the total stomach dry mass was assumed to be grit).

For both goose and ungulate transfer factors (F), it is necessary to estimate the daily dry matter ingestion rate. This can be estimated from body weight (BW, in kg live weight). The hunters were asked to record the weight of the animals, some did and some did not, and so average weights were used where needed. The relationships (from the review of Macdonald 1996) are:

$$\text{Goose ingestion rate (kg d}^{-1}\text{)} = 0.0582 \cdot BW^{0.651}$$

$$\text{Ungulate ingestion rate (kg d}^{-1}\text{)} = 0.0687 \cdot BW^{0.822}$$

2.5.2 Solid/Liquid Partition Coefficients

Aliquots of the soil samples were used to measure solid/liquid partition coefficients (Kd). The Kd is the concentration on the solids divided by the concentration in the pore-water liquids. To obtain pore water, soils were placed in 60 mL syringe tubes (with perforated bottoms, Thibault and Sheppard 1992) and moistened with deionised water until the first drops of drainage occurred. The soils were incubated at room temperature for 7 d, and then were centrifuged at about 6000 m s^{-2} for 1000 s. The extracted pore water was filtered to pass 0.45µm and divided into 2 samples. One was acidified to pH < 2 for analysis of 51 elements by inductively couple plasma mass spectroscopy (ICP-MS), and the other was frozen, not acidified, for analysis by

ICP-MS for I and by instrumental neutron activation analysis (INAA) for Cl. To estimate total concentrations in soil, soil solids were extracted with aqua regia and analysed for the same elements by ICP-MS (see Sheppard et al. 2007 for a discussion of aqua regia for this purpose), except that INAA was used on the whole soil for I and Cl. Values of Kd were computed as:

$$Kd = \frac{C_{solid}}{C_{porewater}} - MC$$

where Kd is in units of L kg^{-1}, C_{solid} is concentration on the dried solids (mg kg^{-1}), $C_{porewater}$ is concentration in the extracted pore water (mg L^{-1}) and MC is the soil moisture content of the solids after extraction but before drying (L kg^{-1}). The correction for moisture content is relevant only for low Kd values numerically in the same range as the moisture content, and accounts for the amount of the element in the residual pore water in the soil when the sample is dried for analysis.

Values of Kd were also obtained for sediment samples. In most cases, it was possible to collect drainage water from the sediment samples as representative of the sediment pore water. In other cases, the water sample from the overlying water was used. The analyses and computations were as for soil Kd.

2.6 QUALITY ASSURANCE

Physical separation was maintained for any samples, such as mineral supplements, that were suspected of having elevated I or trace element concentrations. This included separate drying and grinding equipment, separate time for handling, double bagging of these samples and in most cases separate shipping.

Blind duplicates were prepared from about 10% of the samples. These samples were split from the labelled samples, and the identity of the blind duplicate sample was hidden from the analysts. The laboratory also prepared duplicates and routinely did analysis of certified reference materials along with the samples of this study. Standard reference materials were analysed at the same time as the samples, selecting from the available standard reference materials those that were most like the samples (e.g., reference grain samples to coincide with our plant samples).

Several samples were analyzed to test the potential impact of sampling materials. As described above, some water samples were split between glass and stainless steel shipping containers. In these, it was anticipated that the stainless steel may contaminate the water with metals such as nickel. The knife mills used to grind the samples were steel. To test the impact of this, table sugar samples were sent for analysis, unground and ground. Sugar was chosen because it is a relatively hard solid that should challenge the steel knives of the mill, but is readily soluble so the unground sample was equally soluble for analysis. Although grinding with the mill increased concentrations in sugar of a few metals, such as Cu and Mn, the final concentrations were several orders of magnitude lower than most samples.

2.7 ANALYTICAL METHODS

All analysis of I, Cl and trace elements were done by Activation Laboratories, Ancaster, Ontario. Iodine and trace elements other than Cl in water samples were analysed by inductively coupled plasma – mass spectrometry (ICP-MS). Up to ~60 elements are reported. Care was taken to minimize sample dilution so that the best available detection limit for each element was achieved. Chlorine in water was determined by anion chromatography on an AS9-HC (Anion Separator, column # 9 High Capacity) column and an AG9-HC (Anion Guard column # 9 High Capacity) column followed by suppressed conductivity detection (using an Anion Self Regenerating Suppressor).

Iodine in non-mineral solid samples was determined using alkaline extraction with tetramethylammonium hydroxide (TMAH) (Fecher et al. 1998, DIN EN ISO 17294-1 2005) – "the German Food Standard method – or GFS method". This involves 3 to 4 separate analyses by ICP-MS that include analysis of internal standards, where subsamples are spiked with known amounts of I and analyzed to account for analytical interferences from the sample matrix. Liquid milk samples were also extracted with TMAH and filtered prior to analysis by ICP-MS. Slurry manure was thoroughly shaken, centrifuged and the liquid and solid components were analyzed separately using the methods described above. The two analyses were then used to calculate the initial concentrations in the mixed slurry sample.

Trace elements other than I were analysed in solid non-mineral samples following block digestion[2] of the samples in HNO_3, H_2O_2, HCl and HF.

Iodine in mineral samples (soil and mineral feed supplements) and Cl in both mineral and non-mineral solid samples were analysed by instrumental neutron activation analysis (INAA). In INAA, samples are exposed to a neutron flux in a nuclear reactor, the samples are allowed to rest for a specified time so that short-lived activation products decay away and then the activation products from the analytes of interest are measured by gamma spectroscopy.

Physical properties and pH of soils were measured by ALS Laboratories, Saskatoon, Saskatchewan. Particle size fractionation of the mineral material was into clay, silt and sand fractions by the hydrometer method. Organic carbon was by the Walkeley Black wet oxidation method. Soil pH was determined in water from saturated paste extracts of the soil.

[2] 'block digestion' refers to acid decomposition of a sample in a test tube that is heated by insertion into a close-fitting cylindrical hole in an aluminum block that is heated to a specified temperature. The block ensures uniform heating.

3. RESULTS AND DISCUSSION

3.1 AQUATIC ENVIRONMENT

3.1.1 Fish

The results are presented in a series of tables. Table 3 is the fish/water transfer factor, known by the symbol B, in units L per kg fresh tissue. Data expressed per dry weight of tissue are shown, but are not usually model parameters. The 36 fish sampled include fillets of game fish and whole bodies of smaller fish across 10 lakes. Table 4 has the data for I separated by lake, tissue type and species. Clearly, B values in Lake Sylvia are lower than the other lakes, and the reason for this is not clear. Sylvia Lake does have a high level of suspended and dissolved organic matter content. In general, B values for whole fish were higher than for fillets, but these were also different species. The fillets were mostly of large predators whereas the whole fish were smaller and more likely to include invertebrates and plants in their diet. However, the whole fish samples would have included the thyroid which contains much of the iodine in any species.

Species to species differences, comparing the data for fillets, were not consistent. Sheppard et al. (2006), in a major review of transfer parameters, noted a B value for I of 6 $L\ kg^{-1}$, within 2·GSD of the present number.

The variation in I concentrations from year to year (Figure 3-2), for Northern Pike collected in one lake, varied almost tenfold. There are no water data available for years other than 2008, so it is not possible to determine if this range in tissue concentrations was linked to differences in water concentrations. This is substantial variation and does raise the question of whether water concentrations also fluctuate, and how quickly biota concentrations respond to changes in water concentrations.

The liver and heart tissues results were of a composite of the tissues from 2001 to 2007, and so should be compared to an average of the back muscle concentrations over those years. Both organs appear to have higher I concentrations than the back muscle, which is a common observation for many elements.

Table 3. Fish tissue/water transfer factors (B, computed as concentration ratios). Blank cells indicate the element was not detectable in any samples. Elements are alphabetic after I.

Element	Number of measured fresh weight B	GM of measured fresh weight B (L kg^{-1})	GSD of measured fresh weight B (unitless)	GM of measured dry weight B (L kg^{-1})	GM of water concentration (µg L^{-1})	GM of fresh weight fish tissue concentration (mg kg^{-1})	GM of dry weight fish tissue concentration (mg kg^{-1})
I	36	34	2.7	170	1.4	0.06[a]	0.22
Ag	0						0.0037
Al	0				310		
As	30	7.7	2.8	28	8	0.071	0.26
Au	15	27	1.9	95	0.0059		0.00043
B	0						0.3
Ba	3	4.7	1.3	23	210		5.2
Be	0					0.00021	0.00076
Bi	0						0.004
Br	0				73		
Ca	30	96	4.5	350	9800	810	2900
Cd	24	140	6.1	470	0.018	0.0027	0.0097
Ce	30	3.4	7.5	12	0.27	0.0011	0.0039
Co	30	95	6.3	340	0.061	0.008	0.029
Cr	29	0.28	5.2	1.1	37		0.044
Cs	30	7100	6.9	26000	0.0032	0.026	0.094
Cu	30	150	2.2	580	2	0.35	1.3
Dy	30	4.1	5.5	15	0.017	8.8E-05	0.00032
Er	25	4.3	5.7	16	0.012		0.00022
Eu	21	4.3	4.8	15	0.022		0.00045
Fe	30	33	6.5	120	110	5.4	20
Ga	5	70	4.5	260	0.12		0.023
Gd	8	38	3.8	150	0.03		0.004
Ge	0				0.093		
Hf	7	26	2.6	100	0.041		0.0038
Ho	29	3.4	5.3	12	0.0036	1.6E-05	5.8E-05
In	0				0.001		0.00023
K	30	17000	1.8	58000	220	5500	20000
La	30	4	5.9	14	0.16	0.00079	0.0028
Li	30	3.6	2.8	13	3	0.013	0.047
Lu	17	28	2.6	88	0.0032		0.00033
Mg	30	180	1.8	660	1800	420	1500
Mn	30	130	9.4	480	4.3	0.56	2
Mo	30	8.5	3.9	31	0.46	0.0038	0.014
Na	30	50	2.1	180	7600	520	1900
Nb	18	29	2.6	110	0.0085	0.00042	0.0015
Nd	30	3	7.1	11	0.13	0.00051	0.0018
Ni	6	96	3.7	350	1.1		0.43
Pb	13	47	3	190	0.25		0.043
Pd	0				0.018		
Pr	9	34	6.3	130	0.036		0.0044
Rb	30	69000	3.3	250000	0.15	12	45
Re	2	4.6	4.3	18	0.002		0.0002
Sb	30	11	3.8	40	0.079	0.0011	0.0041
Sc	2	0.62	3.8	2.4	1	0	0.0062
Se	30	10	2.1	37	28	0.35	1.3
Si	0				1900		
Sm	30	5.1	4.7	18	0.03	0.00018	0.00067
Sn	0				0.71		
Sr	30	16	5.7	57	16	0.31	1.1
Ta	9	19	3.2	73	0.0021		0.00029
Tb	29	20	2.9	70	0.0037	9.07E-05	0.00033
Th	4	130	3.5	450	0.012		0.0089
Ti	28	11	8.9	39	4.7		0.23
Tl	16	3300	2.3	10000	0.0026	0.0063	0.023
Tm	7	30	4.5	110	0.002		0.00025
U	8	23	2.1	91	0.066		0.0063
V	15	2.2	6.4	7.2	10		0.071

Element	Number of measured fresh weight B	GM of measured fresh weight B (L kg^{-1})	GSD of measured fresh weight B (unitless)	GM of measured dry weight B (L kg^{-1})	GM of water concentration (µg L^{-1})	GM of fresh weight fish tissue concentration (mg kg^{-1})	GM of dry weight fish tissue concentration (mg kg^{-1})
W	5	150	2.3	380	0.035		0.011
Y	30	3.8	4.8	14	0.13	0.00060	0.0022
Yb	8	24	3.9	92	0.016		0.0018
Zn	30	210	3.3	760	43	8.2	30
Zr	8	21	3	84	1.5		0.13

[a] Northern Pike from ELA had tissue concentrations of: 0.05-0.43 mg kg^{-1} (muscle), 0.56 mg kg^{-1} (liver, composite sample) and 0.42 mg kg^{-1} (heart, composite sample). Lake trout from the same lake had 0.13 mg kg^{-1} (muscle).

Table 4. Variation in fish/water I transfer factors (B, L kg^{-1}) among lakes, with the number of observations in brackets where >1. Blank cells indicate no sample. Samples for ELA, Huron and MacKay were collected in 2008, the remainder in 2007.

Lake	All fillets	Whole fish	Eviscer-ated	Pike fillets	Smallmouth bass fillets	Walleye/ sauger fillets	Other fillets	Macro-phyte[e]
Dog	83 (3)	69[a] 350[b]		83		140	Lake Whitefish 49	200
ELA	18 (2)			8.3			Lake Trout 38	
Huron	27 (5)					19 (2)	Sucker 56 (2) Lake Whitefish 12	
Lac des Milles Lacs	69 (4)			68	39	120	Perch 67	240
MacKay	22 (2)			22			Lake Trout 23	
Sylvia	7.4 (5)			9.2	9.1	6.3 (2)[d]	Sucker 6.7	230
Wabigoon	14 (3)	38[b]		14	15	13		260
Wanapitei	53 (3)	210[c]	65[b]		73	53	Lake Trout 37	450
White		36[b]						160
Whitefish	57 (3)			54	55	61		
Overall	29 (30)	92 (5)	65	26 (7)	29 (5)	28 (9)	31 (9)	280

[a] White Sucker
[b] Yellow Perch
[c] Emerald Shiner
[d] Includes two species commonly called Walleye: *Sander vitreus* and *S. canadensis*
[e] Macrophytes were *Stuckenia* (*Typha* in Dog Lake had B = 58 L kg^{-1})

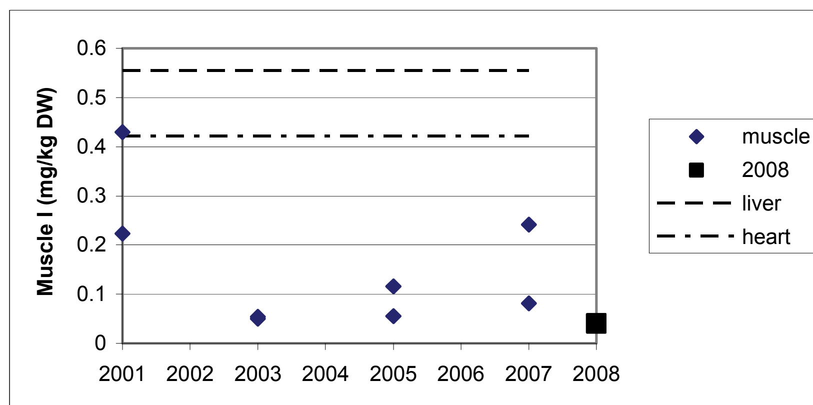

Figure 3-1. Muscle concentrations of I in pike sampled over 7 years at the Experimental Lakes Area, two samples each year except 2008 (the values overlap in 2003). Liver and heart samples were a composite of tissues from 2001 to 2007. No statistically significant correlations were observed with 31 other elements measured in the same tissues.

3.1.2 Macrophyte

A transfer factor from water to macrophytes (Table 5) is not a commonly needed model parameter, but macrophytes do represent primary productivity in water bodies and therefore there is a potential need to estimate impacts to macrophytes. These plants are rooted and will absorb some elements by way of the roots in addition to the exposure to the water column they share with fish. Macrophytes sampled in the same lakes (although not necessarily in the same area in the lakes) had B values that were correlated to those of fish (Figure 3-2).

Table 5. Aquatic macrophyte (*Stuckenia*) tissue/water transfer factors (computed as concentration ratios). Blank cells indicate the element was not detectable in any samples.

Element	Number of measured fresh weight B	GM of measured dry weight B (L kg^{-1})	GSD of measured dry weight B (L kg^{-1})	GM of water concentration (µg L^{-1})	GM of dry weight plant tissue concentration (mg kg^{-1})
I	7	280	1.7	1.4	0.39
Ag	0				0.023
As	7	36	1.8	9.8	0.35
Au	4	880	1.8	0.0044	0.0021
B	0				14
Ba	2	310	1	120	69
Be	0				0.022
Bi	0				0.0076
Br	0			95	
Ca	6	2300	1.5	8500	21000
Cd	5	19000	2.6	0.021	0.47
Ce	7	5600	2	0.3	1.7
Co	7	24000	2.4	0.071	1.7
Cr	7	63	2.9	45	2.8
Cs	7	16000	3.2	0.0038	0.062
Cu	7	4000	1.9	1.9	7.6
Dy	7	3900	2	0.02	0.079
Er	7	3500	2.1	0.013	0.046
Eu	7	1300	2.2	0.028	0.038
Fe	7	8700	2.7	130	1100

Element	Number of measured fresh weight B	GM of measured dry weight B (L kg^{-1})	GSD of measured dry weight B (L kg^{-1})	GM of water concentration (µg L^{-1})	GM of dry weight plant tissue concentration (mg kg^{-1})
Ga	7	1300	3.8	0.14	0.19
Gd	7	3700	2.3	0.034	0.13
Ge	1	870		0.12	
Hf	7	670	4.3	0.046	0.031
Hg	0			0	0.019
Ho	6	3700	2.2	0.005	0.015
In	0				0.0017
K	7	62000	2.5	260	16000
La	7	5500	2.1	0.18	1
Li	7	130	3.5	4.2	0.56
Lu	6	1700	2.2	0.0048	0.0066
Mg	7	1600	1.3	2550	4100
Mn	7	94000	2.3	3.9	370
Mo	7	1000	1.5	0.4	0.4
Na	7	300	2	8860	2700
Nb	4	15000	3	0.014	0.087
Nd	7	5200	2.4	0.15	0.78
Ni	6	4400	2.2	1.3	4.3
Pb	7	3400	2.6	0.22	0.74
Pr	7	5000	2.3	0.04	0.2
Rb	7	89000	3.1	0.17	15
Re	2	43	3.3	0.002	0.000245
Sb	7	170	1.9	0.092	0.016
Sc	3	300	3.7	1	0.17
Sm	7	3900	2.1	0.034	0.13
Sn	5	220	6.8	0.57	0.15
Sr	7	2000	1.4	20	40
Ta	6	2500	12	0.0026	0.0055
Tb	6	3700	2.1	0.0053	0.016
Te	0				0.0058
Th	7	7900	3.2	0.016	0.12
Ti	7	5600	3	6	34
Tl	4	60000	1.8	0.0023	0.13
Tm	6	2700	2.4	0.0026	0.0059
U	7	1600	2.5	0.069	0.11
V	7	160	2.9	13	2
W	5	930	2.9	0.028	0.024
Y	7	3200	2.1	0.14	0.47
Yb	7	2100	2.2	0.02	0.042
Zn	7	800	3.2	47	38
Zr	7	550	4.3	1.9	1

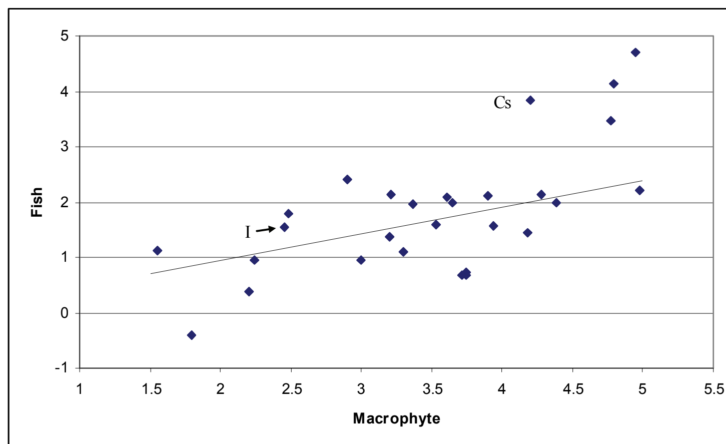

Figure 3-2 Comparison of log fish fresh weight tissue/water concentration ratios (B) versus log macrophyte dry weight tissue/water concentration ratios. Each point is a different element, I and Cs are labelled. The line is the best fit for a constant ratio between the two ratios.

3.1.3 Water Concentrations in 20 Canadian Shield Lakes

A survey of stable element concentrations in water bodies may not have direct implications to environmental risk assessment, but the information is none the less of value. The survey gives an indication of the variability in concentrations to expect among water bodies, which may also apply to radionuclide contaminants. For K, Rb, Th and U, the data can be used directly to compute background concentrations of the corresponding and relevant long-lived radionuclides. For I and Cl, risk assessment of ^{129}I and ^{36}Cl may take into account isotopic dilution from the stable elements. The data obtained are shown in Table 6. The ratio of hypo/meta-limnion concentrations was 0.97 for I and about 1.2 for most other elements and including hardness as $CaCO_3$. This ratio would be greater than unity if meteoric water causes dilution of the metalimnion. In general, and especially for I, these data suggest there is not a large error in fish B values depending on the depth of water sample used. The one very distinct exception was Mn that had up to 350-fold higher concentration in the hypolimnion than the metalimnion (the geometric mean was 43-fold). Although less distinct, the ratio for Fe was also high in some of the same lakes as for Mn. This implies a redox effect, where the hypolimnion was chemically reducing resulting in more Mn^{++} and Fe^{++}, which are more soluble than the oxidized species.

The spatial distribution of lake water (and soil) I concentrations is shown in Table 7. For both, Zones 3 and 4 had low I concentrations. The effect is not large and there is no apparent explanation for why I would be less abundant in these Zones.

Table 6. Water concentrations (μg L^{-1}) 20 lakes, most with hypolimnion and metalimnion sampled separately. Elements with many non-detectable values are not shown. Hardness (mg $CaCO_3$ L^{-1}) is computed from Ca and Mg concentrations

Element (n when N<50)	Average concentration (μg L^{-1})	Standard deviation (μg L^{-1})	Average ratio of hypo-/meta- limnion concentrations	Standard deviation of ratios of hypo-/meta- limnion concentrations
Hardness (mg $CaCO_3$ L^{-1})	32	16	1.2	0.34
I	1.4	0.55	0.97	0.21
Al	310	160	1.1	0.46
As	9.8	7.2	1.2	0.66
Au (20)	0.0073	0.0058	1.8	0.48
Ba (29)	290	190	1.4	0.61
Ca (44)	8470	3920	1.2	0.28
Cd (46)	0.053	0.16	1.5	0.84
Ce	0.28	0.23	1.4	0.72
Co	0.09	0.083	2.6	4.1
Cr	44	29	1.2	0.52
Cs	0.0038	0.0057	1.2	0.61
Cu	2.3	2.7	1.5	1.6
Dy	0.017	0.013	1.2	0.41
Er	0.012	0.0082	1.2	0.3
Eu	0.032	0.043	1.6	1.8
Fe	230	580	5.5	12
Ga	0.14	0.086	1.2	0.44
Gd	0.03	0.025	1.3	0.6
Ge	0.13	0.11	1.4	0.83
Hf	0.037	0.017	1.1	0.68
Ho (47)	0.0037	0.0027	1.2	0.58
K	400	390	1.2	0.79
La	0.17	0.15	1.4	0.55
Li	4.8	3.2	1.2	0.47
Lu (48)	0.0038	0.0042	1.3	0.61
Mg	2140	1140	1.2	0.27
Mn	37	83	43	95
Mo	0.46	0.2	1.5	0.88
Na	10790	6470	1.1	0.61
Nb	0.0084	0.0042	1.1	0.17
Nd	0.15	0.13	1.5	0.72
Ni (46)	1.8	2.8	0.93	0.44
Pb	0.29	0.27	1.9	2.5
Pr	0.039	0.034	1.4	0.63
Rb	0.19	0.17	1.1	0.57
Re (33)	0.011	0.029	1.4	0.63
Sb	0.1	0.057	1.3	0.53
Se	36	26	1.3	0.76
Si	2330	1340	1.5	1.2
Sm	0.031	0.025	1.4	0.47
Sn (44)	0.70	0.70	1.4	1.5
Sr	23	8.9	1.2	0.32
Ta (42)	0.0027	0.0029	1.3	1.1
Tb (42)	0.0040	0.0028	1.1	0.50
Th	0.013	0.014	0.99	0.24
Ti	4.8	2.7	1.2	0.38
Tm (40)	0.0022	0.0014	1.2	0.30
Tl				
U	0.11	0.19	1.1	0.28
V	12	7.7	1.2	0.45
W (41)	0.077	0.15	1.5	1.8
Y	0.13	0.095	1.2	0.39
Yb	0.017	0.014	1.2	0.53
Zn	69	63	1.7	1.6
Zr	1.6	0.66	1.1	0.52

Table 7. Spatial distribution of I concentrations in surface water and soil, showing the results for the sampling zones in Figure 1. Test of significance is for an effect of zone, and both concentrations varied significantly among the zones.

Zone	Surface water (µg L^{-1})	Soils (mg kg^{-1})
1	1.5	15[a]
2	1.2	2.4
3	1.0	2.3
4	1.0	2.0
5	1.4	5.9
Manitoba	1.9	1.2
Significance	$P < 0.005$	$P < 0.001$[a]

[a] There were 2 values for zone 1, 29.9 mg kg^{-1} and <0.5 mg kg^{-1}, the reason for this large difference is not known, when excluded there was no significant difference amount the remaining zones.

3.1.4 Sediment Kd Values

Sediment was collected in the 20 lakes and in several locations or depths in some of these lakes, with the result that there were 33 samples in total. The Kd values for I were among the lowest of the 58 elements shown in Table 8. For I and many of the other elements in Table 8, Kd was notably higher when the water where the sediment was collected was >5 m deep. This separation at 5 m was *a posteriori*, the data appeared to consist of 2 groups above and below this depth. However, there is some mechanistic explanation, because wave action will create a more energetic environment in shallow water which will predispose sediments to be coarser grained and perhaps lower in organic matter content. The effect of depth of water, when it was statistically significant, was typically 4-fold higher in deep-water sediments.

Table 8. Sediment (dry weight) concentration/water concentration partition coefficients (Kd, L kg^{-1}). Blank cells indicate the element was not detectable in any samples.

Element	Number of Kd values	GM of sediment Kd (L kg^{-1})	GSD of sediment Kd (unitless)	GM of dry weight sediment concentration (mg kg^{-1})
I	24	900	4.0	5.1
Ag	1	160		0.022
Al	33	32000	2.1	12000
As	33	1400	4.3	13
B				4.8
Ba	6	110	3.3	100
Be	2	2800	1.6	0.33
Bi				0.15
Ca	26	620	2.3	5000
Cd	31	4000	6.4	0.26
Ce	33	55000	2.5	40
Co	33	22000	3	10
Cr	33	1300	1.7	30
Cs	28	95000	3.5	0.72
Cu	32	5200	3.3	28
Dy	32	49000	2.4	1.8
Er	32	41000	2.3	0.9
Eu	32	11000	1.9	0.57
Fe	30	44000	4.7	21000
Ga	32	35000	2	3.9
Gd	32	37000	2.5	2.5
Ge	26	1300	1.5	0.1
Hf	8	2600	2.7	0.1
Ho	32	48000	2.2	0.33
In	5	53000	1.7	0.033
K	32	920	2.7	1100
La	33	48000	2.6	21
Li	28	3200	2	10
Lu	27	27000	2	0.11
Mg	33	2300	2.3	4700
Mn	33	5300	8	590
Mo	31	870	3.1	0.6
Na	33	57	2.1	800
Nb	10	43000	3.1	0.54
Nd	33	54000	2.7	18
Ni	30	13000	3.7	38
Pb	32	19000	3.2	19
Pr	32	58000	2.7	5.6
Rb	33	14000	3.5	12
Re	14	270	3.4	0.0018
Sb	29	1600	2.1	0.18
Sc	3	3300	1.1	3.1
Se	32	50	2.2	0.9
Sm	32	51000	2.8	3.3
Sn	8	5000	1.7	0.92
Sr	33	860	1.8	27
Tb	32	47000	2.7	0.33
Te	1	200		0.066
Th	32	210000	2.6	4
Tl	20	15000	3.5	0.12
Tm	25	47000	2.3	0.13
U	33	6600	3.3	1.7
V	33	5500	1.8	43
W	12	7600	3.9	0.42
Y	33	42000	2.3	11
Yb	32	26000	2	0.68
Zn	22	380	3.7	68
Zr	33	2300	2.4	3

3.2 TERRESTRIAL ENVIRONMENT – WILD GAME

3.2.1 Geese

The F^g for I in geese not adjusted for grit in the stomach was 4.2, whereas when adjusted for the mass of grit it was 1.1 (Table 9). Similarly, the muscle/stomach concentration ratio CR for I not adjusted for grit in the stomach was 0.54, whereas when adjusted for the mass of grit it was 0.15. It is improbable that geese derive any I from the mineral grit, so that the adjustment for only the mass of the grit is probably appropriate.

The TF values for I, adjusted or not for grit, were much higher than the 0.0087 d kg^{-1} reported by Howard et al. (2009a) for poultry. The TF values for other elements reported for poultry by Howard et al. were also either much higher or much lower than observed here for geese. With the possible exception of Cs, there are few data for birds and the discrepancies noted here indicate the need for further research. This research must address the issue of mineral ingestion, which is present for all animals but is especially important for birds that ingest grit to aid comminution.

Table 9. Goose tissue/stomach content transfer factors (F^g, computed as the fresh weight tissue / dry weight stomach concentration ratio, divided by estimated ingestion rate). Blank cells indicate the element was not detectable in any samples.

Element	Number of measured F^g	GM of measured F^g (d kg^{-1})	GSD of measured F^g (unitless)	F^g corrected for 73% grit (d kg^{-1})	GM of dry weight concentration ratio (unitless)	GM of fresh weight concentration ratio (unitless)	GM of dry weight concentration in stomach content (mg kg^{-1})	GM of fresh weight concentration in muscle (mg kg^{-1})	GM of dry weight concentration in muscle (mg kg^{-1})
I	5	4.2	2.0	1.1	1.9	0.54	0.094	0.051	0.18
Ag	5	0.13	1.5	0.036	0.061	0.017	58	0.44	1.6
As	5	0.66	1.8	0.18	0.3	0.086	0.018		0.001
Au	4	3.7	2.8	1	1.7	0.49	0.077	0.0013	0.0047
B	5	0.15	1.7	0.039	0.067	0.019	0.54	0.046	0.16
Ba	3	0.02	2.4	0.0055	0.0093	0.0027	0.00021	9.7E-05	0.00034
Be	5	0.015	3.5	0.0041	0.007	0.002	6.1	0.12	0.41
Bi	1	0.16		0.042	0.071	0.02	250		2.3
Ca	5	0.19	1.8	0.052	0.088	0.025	0.4	0.00078	0.0028
Cd	5	1.6	1.5	0.44	0.75	0.21	5000	120	440
Ce	5	0.028	2.3	0.0075	0.013	0.0036	0.023	0.0048	0.017
Co	5	0.039	1.6	0.01	0.018	0.005	7.5	0.027	0.095
Cr	5	0.019	1.5	0.0051	0.0087	0.0024	2.6	0.013	0.046
Cs	5	0.43	13	0.12	0.2	0.056	260	0.63	2.2
Cu	5	1.5	1.7	0.4	0.69	0.19	0.62	0.035	0.12
Dy	5	0.032	3.3	0.0087	0.015	0.0042	9.5	1.8	6.5
Er	5	0.029	3.3	0.0078	0.013	0.0038	0.22	0.00092	0.0033
Eu	5	0.024	2.9	0.0063	0.011	0.0031	0.13	0.00048	0.0017
Fe	5	0.14	1.5	0.039	0.066	0.019	0.17	0.00053	0.0019
Ga	4	0.02	1.9	0.0054	0.0094	0.0026	3200	59	210
Gd	5	0.037	2.9	0.0099	0.017	0.0048	5		0.043
Ge	0			0			0.32	0.0015	0.0054
Hf	2	0.048	1.7	0.013	0.023	0.0065	0.2		
Hg	0			0			0.39		0.0089
Ho	5	0.031	3.2	0.0083	0.014	0.004	0.0053		
In	5	0.23	1.6	0.062	0.1	0.03	0.041	0.00016	0.00058
K	5	2.6	1.4	0.7	1.2	0.34	0.0032	9.4E-05	0.00033
La	5	0.027	2.1	0.0072	0.012	0.0035	11000	3600	13000
Li	5	0.061	5.4	0.016	0.028	0.0079	4	0.014	0.049
Lu	2	0.079	2.0	0.021	0.037	0.011	3.4	0.027	0.096
Mg	5	2.3	1.6	0.61	1	0.29	0.018		0.00065
Mn	5	0.059	1.8	0.016	0.027	0.0076	850	250	880
Mo	5	0.078	1.7	0.021	0.036	0.01	3.4	0.034	0.12

Element	Number of measured F^g	GM of measured F^g (d kg^{-1})	GSD of measured F^g (unitless)	F^g corrected for 73% grit (d kg^{-1})	GM of dry weight concentration ratio (unitless)	GM of fresh weight concentration ratio (unitless)	GM of dry weight concentration in stomach content (mg kg^{-1})	GM of fresh weight concentration in muscle (mg kg^{-1})	GM of dry weight concentration in muscle (mg kg^{-1})
Na	5	0.76	1.2	0.21	0.35	0.099	9.6	0.94	3.3
Nb	5	0.034	3.1	0.0093	0.016	0.0044	0.52	0.0023	0.0082
Nd	5	0.034	2.8	0.009	0.015	0.0043	2.6	0.011	0.04
Ni	5	0.023	1.4	0.0062	0.011	0.003	100	0.3	1.1
Pb	5	0.07	2.0	0.019	0.032	0.0091	4.4	0.041	0.14
Pr	5	0.031	2.5	0.0084	0.014	0.004	0.74	0.003	0.011
Rb	5	1.3	1.7	0.34	0.58	0.16	27	4.5	16
Re	5	1.1	2.0	0.3	0.51	0.14	0.0036	0.00052	0.0018
Sb	5	0.27	4.2	0.072	0.12	0.035	0.063	0.0022	0.0077
Sc	5	0.021	4.1	0.0057	0.0097	0.0027	0.44	0.0012	0.0043
Se	0			0				0.19	0.66
Sm	5	0.039	3.0	0.01	0.018	0.005	0.42	0.0021	0.0074
Sn	2	0.45	2.0	0.12	0.22	0.063	0.27		0.067
Sr	5	0.017	2.7	0.0045	0.0076	0.0021	130	0.27	0.95
Ta	2	3.1	12	0.84	1.5	0.42	0.0015	0.00019	0.00066
Tb	5	0.039	3.0	0.011	0.018	0.0051	0.045	0.00023	0.00080
Te	5	1.1	1.5	0.3	0.51	0.14	0.006	0.00088	0.0031
Th	3	0.036	1.4	0.0098	0.017	0.0048	1.4		0.029
Ti	5	0.031	3.6	0.0083	0.014	0.004	120	0.49	1.8
Tl	5	0.27	3.5	0.073	0.12	0.035	0.13	0.0047	0.017
Tm	5	0.025	3.4	0.0069	0.012	0.0033	0.017	5.7E-05	0.00020
U	5	0.019	2.1	0.0052	0.0088	0.0025	0.44	0.0011	0.0038
V	5	0.032	2.9	0.0086	0.015	0.0041	5.4	0.022	0.079
W	3	0.056	3.6	0.015	0.026	0.0074	0.33		
Y	5	0.029	3.2	0.0078	0.013	0.0037	1.2	0.0044	0.015
Yb	5	0.023	3.5	0.0061	0.01	0.0029	0.12	0.00037	0.0013
Zn	5	31	1.8	8.5	14	4.1	8.1	33	120
Zr	3	0.022	2.8	0.0059	0.0099	0.0028	14		0.15

3.2.2 Deer and Other Ungulates

The data for deer are shown in Table 10, with the transfers computed as F^d (d kg^{-1}) and as CR values on both tissue dry weight and wet weight bases. The F^d for I is 0.0051 d kg^{-1} compared to 0.0067 d kg^{-1} for beef and 0.041 d kg^{-1} for pig from Howard et al. (2009a) and 0.012 d kg^{-1} for generic animals from Sheppard et al. (2006). The corresponding fresh weight CR values are for 0.013 for deer (this study), 0.095 for beef and 0.093 for pig (Howard et al. 2009b). All of these are in reasonable agreement.

Tables 11 and 12 give F^d and fresh weight CR values for deer, caribou, elk and moose. For I, these values are in good agreement with those of Howard et al. (2009a,b). In terms of total I in the diet, it is probable that domestic animals receive more I because of mineral supplements. Values of CR for Ca, Cd, Co, Mo, Na and U were markedly higher in domestic animals (based on Howard et al. 2009b) than observed here in deer, caribou, elk and moose. Of these, Ca, Co, Mo and Na will be added in at least some domestic feed as mineral supplements. Table 13 is the concentrations in lichen and droppings as used for caribou substrate.

The CR values for ungulates were for lean muscle. In one deer, a fat sample was retained. It had 0.006 mg I kg dry weight compared to 0.01 mg I kg dry weight muscle and 0.004 mg I kg fresh weight muscle from the same animal, not markedly different.

Table 10. Deer tissue/stomach content transfer factors (F^d, computed as fresh weight tissue / dry weight stomach concentration ratio, divided by estimated ingestion rate). Blank cells indicate the element was not detectable in any of the corresponding samples.

Element	Number of measured F^d	GM of measured F^d (d kg^{-1})	GSD of measured F^d (unitless)	GM of dry weight concentration ratio (unitless)	GM of fresh weight concentration ratio (unitless)	GM of dry weight concentration in stomach content (mg kg^{-1})	GM of fresh weight concentration in muscle (mg kg^{-1})	GM of dry weight concentration in muscle (mg kg^{-1})
I	13	0.0051	2.0	0.045	0.013	0.41	0.0054	0.019
Ag	12	0.028	4.8	0.25	0.073	140	0.18	0.64
As	13	0.015	1.5	0.13	0.039	0.0015		0.0046
Au	1	0.99		9	2.7	0.0093		0.0025
B	13	0.0026	1.9	0.023	0.0066	1.3	0.049	0.17
Ba	0					1E-04		0.00027
Be	13	0.0072	1.5	0.064	0.018	34	0.22	0.77
Bi	4	0.28	4.4	2.8	0.78	54		
Ca	13	0.0012	1.8	0.01	0.003	0.0051	9.5E-05	0.00033
Cd	13	0.0085	1.8	0.076	0.022	16000	47	160
Ce	13	0.0036	2.2	0.032	0.0092	0.15	0.0033	0.011
Co	13	0.0038	2.4	0.034	0.0099	0.19	0.0017	0.0059
Cr	13	0.038	2.1	0.34	0.097	0.26	0.0026	0.0089
Cs	13	0.20	2.2	1.7	0.50	0.47	0.045	0.16
Cu	13	0.12	1.7	1.1	0.32	0.074	0.037	0.13
Dy	13	0.0028	1.8	0.025	0.0072	6.4	2	7
Er	10	0.0032	1.7	0.029	0.0084	0.0075	5.5E-05	0.00019
Eu	9	0.0022	1.8	0.021	0.0061	0.0041		0.00012
Fe	13	0.12	1.6	1	0.30	0.0086		0.00017
Ga	0					140	41	140
Gd	1	0.0058		0.053	0.016	0.028		
Ge	0					0.014		0.001
Hg	10	0.046	1.3	0.4	0.12	0.0042		
Ho	13	0.0031	1.6	0.027	0.0079	0.036		0.015
In	10	0.074	3.2	0.65	0.19	0.0014	1.1E-05	3.8E-05
K	13	0.10	1.5	0.89	0.26	0.00037		0.00025
La	13	0.0026	2.0	0.023	0.0068	16000	4100	14000
Li	13	0.0071	2.5	0.063	0.018	0.13	0.00090	0.0031
Lu	0					5	0.092	0.32
Mg	13	0.045	2.0	0.4	0.12	0.0012		
Mn	13	0.00052	1.8	0.0046	0.0013	2300	260	910
Mo	13	0.0024	3.4	0.021	0.0061	0.86	0.0052	0.018
Na	13	0.013	1.4	0.11	0.032	18	0.57	2
Nb	12	0.0074	1.7	0.066	0.019	0.015		0.001
Nd	13	0.0031	2.3	0.027	0.0079	0.078	0.00062	0.0021
Ni	6	0.027	1.9	0.22	0.065	1.1		0.28
Pb	13	0.042	27	0.37	0.11	0.3	0.032	0.11
Pr	9	0.0048	1.6	0.046	0.013	0.021		0.00098
Rb	13	0.098	1.8	0.87	0.25	24	6.1	21
Re	4	0.0049	1.4	0.049	0.014	0.0014		0.0003
Sb	13	0.088	11	0.79	0.23	0.012	0.0028	0.0096
Sc	2	0.0056	1.8	0.066	0.019	0.015		0.001
Se	10	0.16	1.8	1.4	0.40	0.37	0.15	0.53
Sm	13	0.0052	1.5	0.046	0.013	0.013	0.00017	0.00058
Sn	0					0.07		0.84
Sr	13	0.00026	1.7	0.0024	0.00068	43	0.029	0.1
Ta	8	0.012	1.6	0.11	0.031	0.001		0.00012
Tb	13	0.0038	1.5	0.034	0.0099	0.0018	1.8E-05	6.1E-05
Te	13	0.055	2.0	0.49	0.14	0.0057	0.0008	0.0028
Th	0					0.018		
Ti	13	0.007	4.3	0.063	0.018	3.6	0.065	0.22
Tl	6	0.014	2.0	0.13	0.037	0.0088		0.0013
Tm	0					0.00056		
U	1	0.0093		0.091	0.024	0.011		0.002
V	4	0.010	6.0	0.1	0.029	0.2		0.024
W	1	0.036		0.24	0.071	0.011		0.007
Y	13	0.0037	1.5	0.033	0.0095	0.046	0.00044	0.0015
Yb	0					0.0047		
Zn	13	0.38	1.4	3.4	0.98	42	41	140
Zr	0					0.12		

Table 11. Ungulate transfer factors ($F^{ungulate}$, d kg^{-1}) for selected elements computed as fresh weight tissue / dry weight substrate concentration ratio, divided by estimated ingestion rate). For deer, elk and moose, the substrate concentrations were stomach contents when the animals were killed. For caribou, the substrate was the average concentration in nearby lichen and faecal samples. There was one sample each of caribou, elk and moose.

Element	Deer	Caribou	Elk	Moose	Element	Deer	Caribou	Elk	Moose
I	0.0051	0.0029	0.0020	0.010	Mo	0.0024	0.0048	0.00026	0.0022
As	0.015	0.0084	0.041	0.019	Na	0.013	0.077	0.0062	0.0040
Ca	0.0012	0.0031	0.00042	0.00078	Nb	0.0074	0.00035		0.0083
Cd	0.0085	0.0054	0.0010	0.0030	Nd	0.0031	0.00002	0.00015	0.0010
Ce	0.0036	0.00036	0.00017	0.00091	Ni	0.027			0.012
Cl					Pb	0.042			1.6
Co	0.0038	0.0032	0.0023	0.0090	Rb	0.098	0.90	0.033	0.069
Cr	0.038	0.00025	0.0061	0.020	Sb	0.088	0.0032	0.0068	0.029
Cs	0.20	0.89	0.37	0.14	Se	0.16	0.30	0.20	0.075
Cu	0.12	0.10	0.018	0.023	Sr	0.00026	0.00040	0.00010	0.00010
Fe	0.12	0.0064	0.024	0.098	Th				
K	0.10	0.40	0.026	0.037	Tl	0.007	0.011		
La	0.0026	0.00072	0.00029	0.0014	U	0.0093	0.00059		
Mg	0.045	0.074	0.024	0.035	V	0.010			
Mn	0.00052	0.00066	0.00014	0.00009	Zn	0.38	0.40	0.12	0.12

Table 12. Ungulate muscle/substrate concentration ratios (CR) for selected elements computed as fresh weight tissue / dry weight substrate concentration). For deer, elk and moose, the substrate concentrations were stomach contents when the animals were killed. For caribou, the substrate was the average concentration in nearby lichen and faecal samples. There was one sample each of caribou, elk and moose.

Element	Deer	Caribou	Elk	Moose	Element	Deer	Caribou	Elk	Moose
I	0.013	0.010	0.014	0.088	Mo	0.0061	0.017	0.0018	0.01931
As	0.039	0.029	0.28	0.16	Na	0.032	0.27	0.043	0.035
Ca	0.003	0.011	0.0029	0.0068	Nb	0.019	0.0012		0.072
Cd	0.022	0.019	0.0072	0.026	Nd	0.0079	6.78E-05	0.0010	0.0089
Ce	0.0092	0.0013	0.0012	0.0079	Ni	0.065			0.10
Cl					Pb	0.11			
Co	0.0099	0.011	0.015	0.079	Rb	0.25	3.2	0.23	0.60
Cr	0.097	0.00089	0.042	0.17	Sb	0.23	0.011	0.047	0.25
Cs	0.50	3.1	2.6	1.2	Se	0.40	1.0	1.4	0.65
Cu	0.32	0.35	0.12	0.20	Sr	0.00068	0.0014	0.00071	0.00088
Fe	0.30	0.022	0.16	0.86	Th				
K	0.26	1.4	0.18	0.32	Tl	0.037	0.040		
La	0.0068	0.0025	0.0020	0.012	U	0.024	0.0021		
Mg	0.12	0.26	0.17	0.30	V	0.029			
Mn	0.0013	0.0023	0.0010	0.00077	Zn	0.98	1.4	0.83	1.0

Table 13. Concentrations (μg kg^{-1}) measured in one lichen and one sample of caribou scat from the MacKay Lake region, NWT.

Element	Lichen	Caribou scat	Element	Lichen	Caribou scat	Element	Lichen	Caribou scat
I	1110	629	Ga	1570	567	Sb	83.6	14.8
Ag	167	148	Gd	288	117	Sc	748	279
As	1190	365	Hf	206	67	Se	200	200
Au	0.9	0.2	Hg	137	104	Sm	458	165
B	2900	2800	Ho	37.7	14.9	Sn	180	60
Ba	86000	68000	In	4.1	1.5	Sr	38700	23100
Be	213	80.1	K	3410000	1830000	Ta	28.4	9.4
Bi	39	17	La	2810	1070	Tb	62.7	24.4
Ca	3110000	3280000	Li	4310	1430	Te	6	5
Cd	167	308	Lu	12.7	5.9	Th	631	217
Ce	5860	2080	Mg	1100000	834000	Ti	166000	57000
Cl	200,000	< 100,000	Mn	109000	209000	Tl	68	36.6
Co	2290	1270	Mo	174	238	Tm	12.9	5.53
Cr	14800	2310	Na	2270000	850000	U	186	86
Cs	756	464	Nb	378	122	V	6610	2420
Cu	7330	7110	Nd	2580	923	W	154	38
Dy	215	83.7	Ni	10500	4400	Y	1050	393
Er	125	52.5	Pb	3980	1530	Yb	89.6	41.7
Eu	128	58.5	Pr	650	233	Zn	37800	52500
Fe	2800000	1030000	Rb	13600	8790	Zr	7780	2280

3.3 TERRESTRIAL ENVIRONMENT - LIVESTOCK

3.3.1 Dairy

Iodine and Cl were detectable in all milk and dairy-feed samples, and the resulting F^{milk} values are shown in Table 14. For I, the values are 6-fold higher than from the review by Sheppard et al. (2006 and references cited therein). This is within 2·GSD, so are not considered statistically different. The values of Howard et al. (2009a) are 8-fold lower than those measured here, but their values are based largely on short-term exposures with ^{131}I which may not be in isotopic equilibrium with the full body I load. Conversely, there was potential for residual I from previous teat dips to bias the values observed here upward. The F^{milk} values for Cl were within 4-fold of the review (Sheppard et al. 2006), again not statistically different. Most other elements are in very good agreement between Howard et al. (2009a) and this study (Figure 3-3). Some of the exceptions, such as Nb and U, were cases where our detection limits were limiting the number of observations, and so our results are less useful. With the exception of a few elements such as Cs, I and Sr, the present study has often more than doubled the numbers of values reported by Howard et al. (2009a), substantially improving confidence in milk transfer factors.

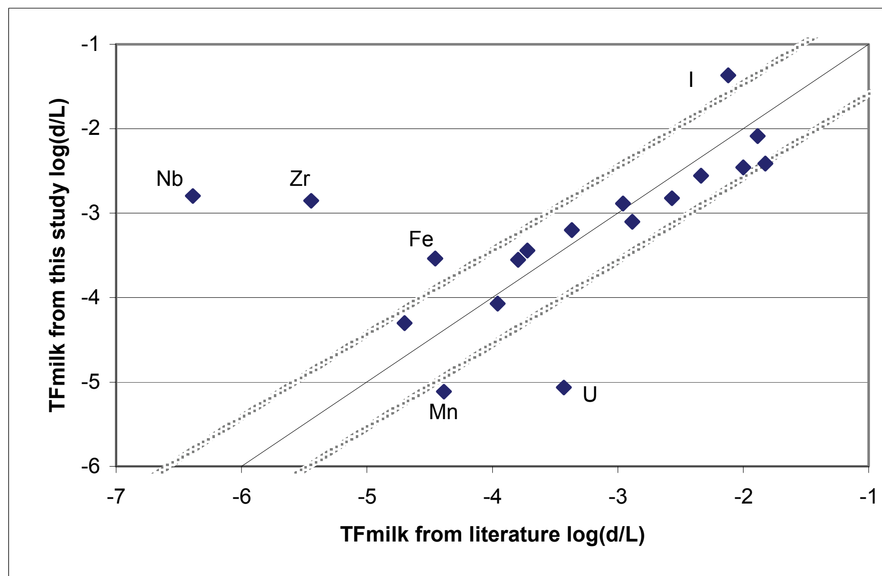

Figure 3-3. F^{milk} measured using stable elements in this study versus those from the literature (Howard et al. 2009, Sheppard et al. 2006). The black line is the 1:1 line, the grey lines are 2·GSD above and below. The elements not labelled, i.e. those that do not differ statistically between this study and the literature, are Cl, Ba, Ca, Ce, Co, Cr, Cs, Mo, Na, Pb, Sr and Zn.

Table 14. Feed-to-milk transfer factors, F^{milk}, (d L^{-1}) from individual cows, sampled from 6 herds, twice each herd. Where N<12, the element was not detectable in all of the 12 samples.

Element	N	GM	GSD	Sheppard et al. 2006	Howard et al. 2009a
I	12	0.043	3.1	0.0076	0.0054
Cl	12	0.0039	2.4	0.015	
As	3	0.00048	1.4		
Au	10	0.077	3.6		
Ba	12	0.00028	1.8		0.00016
Bi	2	0.34	8.2		
Ca	1	0.0035	---		0.010
Ce	7	0.000050	1.5		0.00002
Co	12	0.000085	2.3		0.00011
Cr	4	0.00063	1.5		0.00043
Cs	12	0.0028	1.5		0.0046
Cu	12	0.00012	1.9		
Fe	7	0.00029	2.0		0.000035
Ga	12	0.0057	1.7		
Hf	12	0.0027	1.9		
La	4	0.000050	2.6		
Mg	12	0.0016	1.5		
Mn	12	0.0000077	1.7		0.000041
Mo	12	0.0013	2.2		0.0011
Na	12	0.0082	2.4		0.013
Nb	12	0.0016	1.9		0.00000041
Nd	4	0.000031	2.0		
Pb	9	0.00036	3.8		0.00019
Rb	12	0.0061	1.4		
Re	6	0.0072	7.6		
Se	11	0.0031	2.0		
Sr	12	0.00079	1.6		0.0013
Ta	1	0.00049	---		
Th	12	0.00013	1.7		
Ti	12	0.014	1.6		
Tl	5	0.00044	1.3		
U	1	0.0000086	---	0.00037	0.0018
Zn	12	0.0015	1.7		0.0027
Zr	12	0.0014	1.8		0.0000036

3.3.2 Eggs

Iodine and Cl were detectable in all egg and layer-feed samples, and the resulting F^{egg} values are shown in Table 15. For I, the values are 3-fold higher than from the review by Sheppard et al. (2006 and references cited therein). This is just within 2·GSD, so are not considered statistically different. There is no known potential bias in the observed values. In general, the GSD values are low relative to other transfer parameters, and this reflects the uniformity of egg production farming. The feeds are nearly all commercially supplied to meet standards common across Canada, the genetic background of the birds is nearly all traceable to one breeding facility in Canada, probably all because the eggs produced must be of standard size and quality to enter the market. Comparison of the F^{egg} values measured in this study with those of Howard et al. (2009a) again shows general agreement (Figure 3-4), and in most cases there were more data from the present study than reported by Howard et al. (2009a).

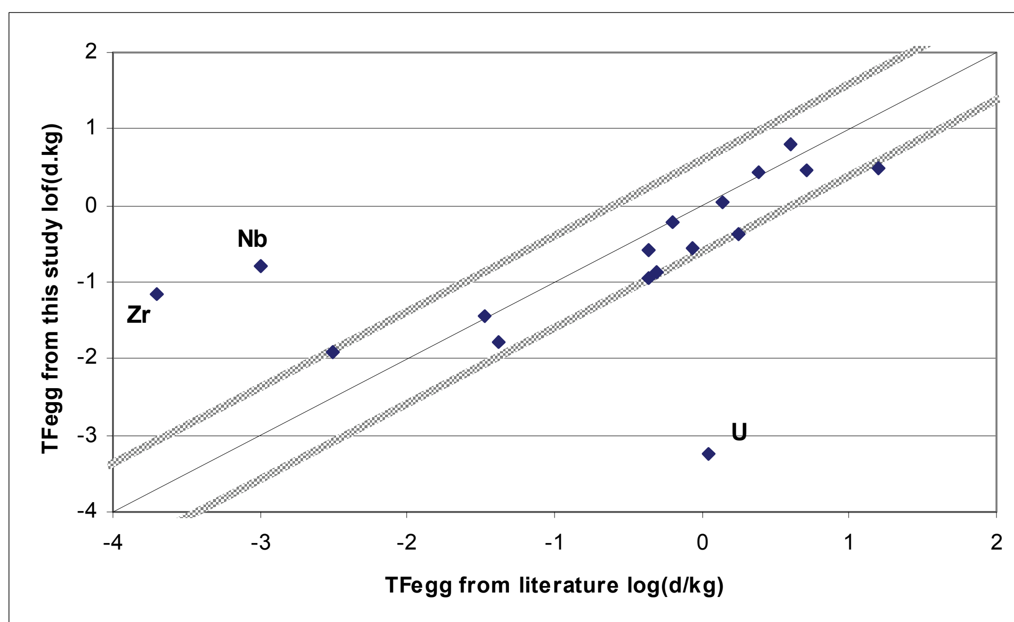

Figure 3-4. F^{egg} measured using stable elements in this study versus those from the literature (Howard et al. 2009, Sheppard et al. 2006). The black line is the 1:1 line, the grey lines are 2·GSD above and below. The elements not labelled, i.e. those that do not differ statistically between this study and the literature, are Ba, Ca, Ce, Co, Cs, Fe, I, Mn, Mo, Na, Se, Sr, Te and Zn.

Table 15. Feed-to-egg transfer factors, F^{egg}, (d kg^{-1}) sampled from 6 flocks, twice each flock. Where N<12, the element was not detectable in all of the 12 samples.

Variable	N	GM	GSD	Sheppard et al. 2006	Howard et al. 2009a
I	12	2.7	1.7	7.5	2.4
Ag	7	0.19	1.9		
As	8	0.38	2.4		
B	12	0.22	1.4		
Ba	2	0.35	1.3		0.87
Be	5	0.0078	1.9		
Bi	2	0.74	1.5		
Ca	12	0.11	1.4		0.44
Cd	12	0.067	2.2		
Ce	4	0.015	1.8		0.0031
Cl	12	6.0	1.5	2.0	
Co	12	0.035	2.1		0.033
Cr	8	0.033	3.1		
Cs	12	0.26	2.1		0.40
Cu	12	0.44	2.1		
Dy	9	0.010	9.1		
Eu	11	0.051	2.0		
Fe	12	0.43	1.4		1.8
Ga	12	0.041	2.5		
Ho	10	0.0051	3.9		
In	4	0.21	1.2		
K	12	2.0	1.7		
La	3	0.046	8.1		
Li	12	0.99	2.2		
Mg	12	0.61	1.3		
Mn	12	0.016	1.6		0.042
Mo	12	0.60	2.0		0.64
Na	12	6.3	1.4		4.0
Nb	2	0.30	3.2		0.001
Nd	10	0.0068	4.9		
Pb	1	0.055	---		
Pr	1	0.073	---		
Rb	12	2.5	1.4		
Re	11	1.5	1.9		
Sb	9	0.079	2.3		
Se	11	3.0	1.6		
Sm	8	0.023	7.2		
Sr	12	0.14	1.6		0.49
Ta	9	0.19	2.3		
Tb	11	0.013	2.4		
Te	4	2.8	2.1		5.1
Ti	3	0.072	13.1		
Tl	12	1.0	2.7		
U	1	0.024	---	1.2	1.1
V	2	0.16	13.8		
Y	12	0.0031	3.3		
Zn	12	1.1	1.3		1.4
Zr	10	0.068	1.5		0.0002

3.3.3 Meat Birds (Chicken, Squab, Turkey, Goose)

The data for transfer to chicken meat, F^c, and the muscle/feed concentration ratios are shown in Table 16. These data combine chickens from very small Cornish hens (~! Kg live weight) to large roasting chickens (~3.6 kg live weight): no effect of body size was evident in the data. Similarly, the white and dark meat of each bird was sampled separately, but they did not different in elemental concentration and so these concentrations were average for each bird.

There were large differences between the values here and those reported by Howard et al. (2009a), and typically data for poultry are scarce and variable. Compared to the F^c for I here of 0.18 d kg^{-1}, Howard et al. (2009a) reported 0.0087 d kg^{-1} based on 3 values and Sheppard et al. (2006) reported 7.5 d kg^{-1} based on 1 value for meat and 2 for eggs. The present data substantially increases the amount of data available.

Perhaps the most interesting comparisons for meat birds are among chicken, goose, squab and turkey (Tables 17 and 18). The F values for squab are often ~100-fold higher than for the other species, whereas the CR values are more consistent across species. This effect of body size is more evident for meat animals, discussed in the next section.

Table 16. Feed-to-meat transfer factors for chicken including Cornish, F^c, (d kg^{-1}) and muscle/feed concentration ratios (CR) sampled from 8 flocks. White and dark meat concentrations were averaged because they were not consistently different. Where N<8, the element was not detectable in all of the 8 samples.

Element	N	GM of F^c	GSD of F^c	GM of CR	GSD of CR
I	8	0.18	4.6	0.018	4.6
Ag	8	0.31	2.3	0.03	2.2
As	7	0.35	3.5	0.033	3.9
Au	1	2.4	---	0.14	---
B	6	0.24	2.7	0.025	2.2
Be	6	0.034	4.5	0.0031	4.6
Bi	3	1.2	3.7	0.15	3.9
Ca	8	0.065	1.6	0.0064	1.6
Cd	8	0.15	2.1	0.015	2.1
Ce	7	0.020	4.3	0.0019	4.0
Cl	8	2.1	2.2	0.21	2.0
Co	8	0.041	2.2	0.0040	2.2
Cr	8	0.048	7.1	0.0047	7.9
Cs	8	1.9	1.9	0.19	1.9
Cu	8	0.28	3.4	0.028	3.0
Dy	8	0.012	3.0	0.0012	3.2
Er	7	0.011	2.6	0.0012	2.6
Eu	8	0.029	6.2	0.0028	7.0
Fe	8	0.20	2.2	0.020	2.2
Ga	8	0.041	3.1	0.0041	3.1
Ge	1	3.0	---	0.40	---
Ho	8	0.0072	2.8	0.000718	3.4
In	6	0.66	1.9	0.072	2.0
K	8	4.2	2.2	0.41	1.7
La	6	0.031	1.9	0.0029	1.5
Li	5	0.15	3.7	0.016	3.2
Lu	2	0.5	1.3	0.044	1.4
Mg	8	1.0	1.8	0.10	1.9
Mn	8	0.017	1.9	0.0017	1.7
Mo	8	0.23	1.9	0.023	1.6

Element	N	GM of F^c	GSD of F^c	GM of CR	GSD of CR
Na	8	4.4	1.9	0.43	1.9
Nb	6	0.047	2.2	0.0052	1.9
Nd	6	0.018	3.2	0.0020	3.1
Ni	2	0.42	1.0	0.053	1.1
Pb	6	0.39	2.4	0.041	2.0
Pr	3	0.069	1.9	0.0067	1.8
Rb	8	8.9	1.5	0.89	1.4
Re	2	0.13	1.5	0.018	1.5
Sb	6	0.093	3.1	0.010	2.6
Sc	2	0.091	1.3	0.0081	2.3
Se	7	8.8	2.1	0.83	1.6
Sm	8	0.020	3.5	0.0020	3.9
Sn	4	1.9	1.4	0.19	1.9
Sr	8	0.039	1.9	0.0038	1.8
Ta	4	0.11	1.5	0.014	1.5
Tb	8	0.051	4.5	0.005	4.3
Te	4	1.2	1.8	0.13	1.8
Ti	6	0.019	3.5	0.0019	3.7
Tl	8	1.7	2.2	0.17	2.6
Tm	3	0.052	3.8	0.0051	4.1
U	2	0.012	2.1	0.0010	1.2
V	1	0.028	---	0.0016	---
W	1	1.2	---	0.11	---
Y	8	0.007	2.9	0.00069	3.4
Yb	2	0.037	3.0	0.0031	1.8
Zn	8	1.0	1.6	0.10	1.4
Zr	7	0.074	2.2	0.0071	2.1

Table 17. Poultry feed-to-meat transfer factors (F^{bird}, d kg^{-1}) for selected elements computed as fresh weight tissue / dry weight feed concentration ratio, divided by estimated ingestion rate).

Element	Chicken	Goose	Squab	Turkey	Element	Chicken	Goose	Squab	Turkey
I	0.18	4.2	25	0.036	Mo	0.23	0.59	10	0.69
As	0.35	0.79	0.8	2.4	Na	4.4	7.6	47	1.2
Ca	0.065	0.24	1.5	0.018	Nb	0.047			
Cd	0.15	0.25	1.6	0.13	Nd	0.018			
Ce	0.02	0.23	2.2	0.041	Ni	0.42			
Cl	2.1	2.8	22	0.58	Pb	0.39	0.21		
Co	0.041	0.23	2.4	0.051	Rb	8.9	4.6	28	5.2
Cr	0.048	0.36	4.3	0.016	Sb	0.093		7.2	0.097
Cs	1.9	8.5	34	2.3	Se	8.8		6.9	2
Cu	0.28	4.6	17	0.068	Sr	0.039	0.059	0.35	0.0038
Fe	0.2	4.0	25	0.13	Th				
K	4.2	4.0	27	2.9	Tl	1.7	2.2	59	0.19
La	0.031	0.35	2.9	0.061	U	0.012			
Mg	1.0	0.96	5.3	1.1	V	0.028		1.6	
Mn	0.017	0.059	0.38	0.0054	Zn	1	2.1	9.3	0.32

Table 18. Poultry feed-to-meat concentration ratio (CR) for selected elements computed as fresh weight tissue / dry weight feed concentration ratio.

Element	Chicken	Goose	Squab	Turkey	Element	Chicken	Goose	Squab	Turkey
I	0.018	0.76	0.76	0.006	Mo	0.023	0.10	0.300	0.12
As	0.033	0.14	0.024	0.41	Na	0.43	1.4	1.4	0.20
Ca	0.0064	0.042	0.044	0.003	Nb	0.0052			
Cd	0.015	0.045	0.047	0.022	Nd	0.002			
Ce	0.0019	0.04	0.065	0.0069	Ni	0.053			
Cl	0.21	0.50	0.67	0.097	Pb	0.041	0.037		
Co	0.004	0.041	0.071	0.0085	Rb	0.89	0.83	0.82	0.88
Cr	0.0047	0.064	0.13	0.0027	Sb	0.01		0.22	0.016
Cs	0.19	1.5	1.0	0.38	Se	0.83		0.21	0.34
Cu	0.028	0.82	0.51	0.011	Sr	0.0038	0.011	0.011	0.00064
Fe	0.02	0.72	0.75	0.021	Th				
K	0.41	0.71	0.80	0.49	Tl	0.17	0.39	1.8	0.031
La	0.0029	0.062	0.087	0.01	U	0.0051			
Mg	0.10	0.17	0.16	0.18	V	0.001			
Mn	0.0017	0.01	0.011	0.00090	Zn	0.10	0.38	0.28	0.053

3.3.4 Meat Animals (Beef, Pork, Lamb, Rabbit)

Measuring transfers to beef meat is much more complicated than most other domestic species because beef in particular often have free access to feed and mineral-enriched salt. However, close to slaughter they are typically kept in feedlots, so that the feed materials and total intake for the herd are well defined. The manure/feed CRs were especially important to assure the feed concentration data, and the median for all elements and farms was 3.6. As with dairy, the manure/feed ratios were low for Cl (0.64), K (0.43) and Rb (0.37). One farm had manure/feed ratios that were notably high (>50) for a number of metals including Ca, Cs, La, Li, Ti, Sm and Zr. Several of these elements are typcially more abundant in soil than feed, and it is probable the manure sample from the feedlot on this farm included some soil.

Both F^{beef} and CR and are shown in Table 19. The median GSD for CR was 2.6 compared to 3.0 for F^{beef}. The computation of F^{beef} accounts for effects of animal size and feed intake and use of TF in assessment models was probably intended to lessen variability in the data. However, it would appear that at least for indigenous elements in beef, F^{beef} values are more variable than the underlying CR values. This trend was also somewhat evident for pig (Table 20), where the median GSD for CR was 2.1 compared to 2.3 for F^{pig}.

Comparison of values here to those in the literature (Figure 3-5), shows similar agreement (and disagreement) as discussed previously for milk and egg.

There was one liver sample each for beef and pig. The F^{beef} for I in liver was 0.0051 d kg^{-1} compared to 0.0024 d kg^{-1} for muscle of the same animal, a difference that is less than 2·GSD. Other elements were more different, the notable liver/muscle concentration ratios were Cd: 20, Co: 10, Cu: 110, Mn: 12 and Mo: 80. With the exception of Cd, these are essential elements. The F^{pig} for liver was from a different animal than the muscle sample, and were 0.0057 d kg^{-1} for liver compared to 0.0019 d kg^{-1} from pig muscle on the same farm. Many of the other elements had liver/muscle ratios above 10, most notably Cs: 2300, K: 110, Rb: 58 and Se: 37000. These liver/muscle ratios are sufficiently high that consumption of offal may be an important pathway relative to muscle meat for radioisotopes of Cs and Se (note the ratio in beef was only 3 for Cs and Se was not detectable in both tissues).

As with poultry, the comparisons among beef, pig, lamb and rabbit (Tables 21 and 22) is instructive. Considering I, the F values increased 88-fold from beef to rabbit, inversely proportional to body mass as expected. In contrast, the CR values were within twofold across the species. This trend was evident for the most of the elements, whether essential or not. This is consistent with Howard et al. (2009b) who conclude that CR is perhaps a better model of feed-to-meat transfer than is F.

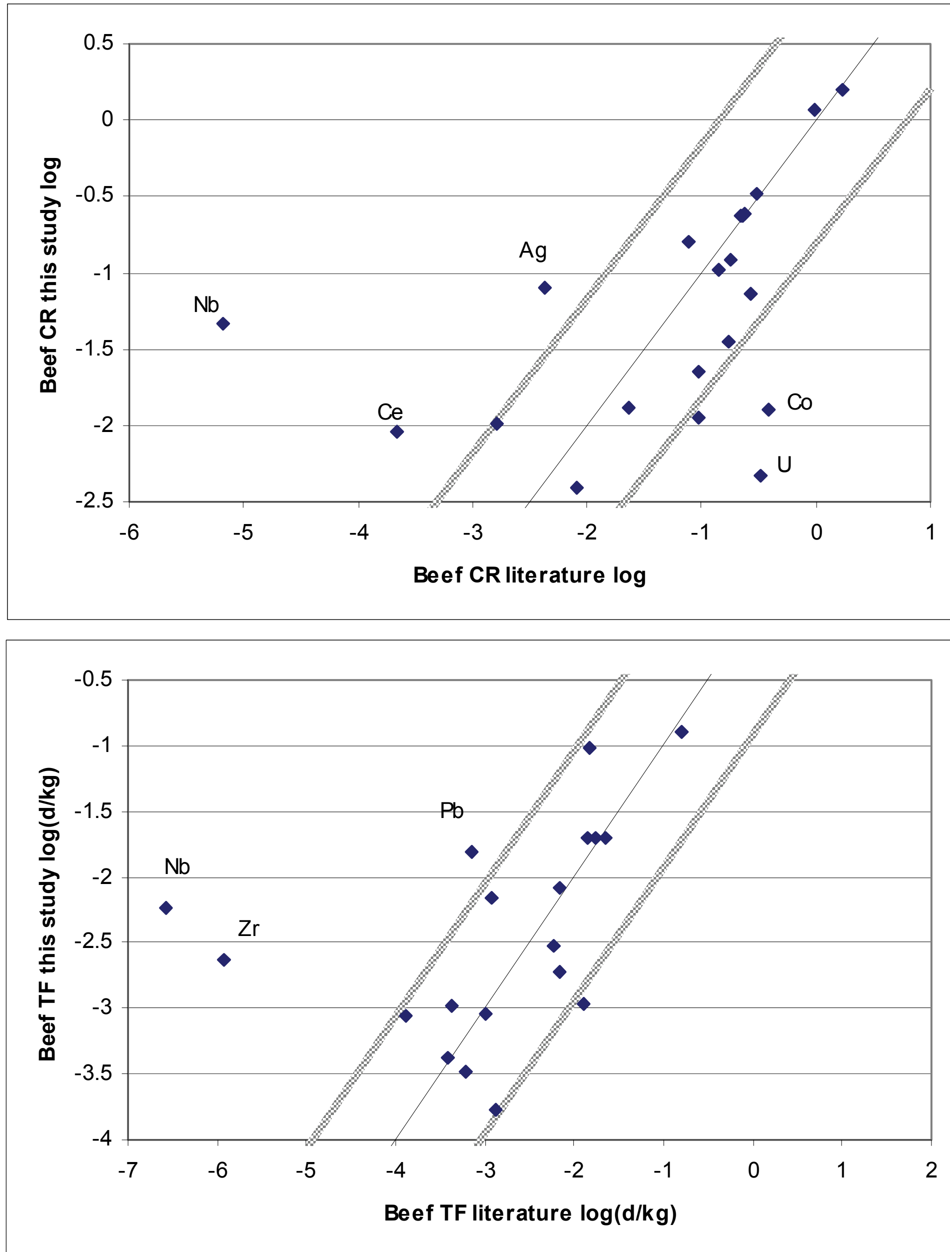

Figure 3-5. CR for beef and F^{beef} measured using stable elements in this study versus those from the literature (Howard et al. 2009, Sheppard et al. 2006). The black line is the 1:1 line, the grey lines are 2·GSD above and below. The elements not labelled for CR_{beef}, i.e. those that do not differ statistically between this study and the literature, are I, Cl, Ca, Cd, Cs, Fe, La, Mg, Mn, Mo, Na, Pb, Rb, Sb, Te and Zn, and those for TF_{beef} are I, Cl, Ca, Cd, Co, Cs, Fe, La, Mn, Mo, Na, Sb, Sr, Te, U and Zn.

Table 19. Feed-to-meat transfer factors for beef, F^{beef}, (d kg^{-1}) and muscle/feed concentration ratios (CR) sampled from 16 animals. Where N<16, the element was not detectable in all of the 16 samples.

Element	N	GM of F^{beef}	GSD of F^{beef}	GM of CR	GSD of CR
I	16	0.0018	3.3	0.022	3.3
Ag	7	0.0082	2.1	0.096	1.8
As	13	0.011	2.6	0.14	2.6
Au	4	0.059	3.1	0.69	3.6
B	13	0.0033	2.6	0.042	2.8
Be	7	0.0014	2.5	0.017	2.2
Bi	2	0.0056	2.4	0.081	2.1
Ca	16	0.0011	1.9	0.013	1.8
Cd	16	0.0025	2.9	0.031	2.6
Ce	12	0.00078	4.6	0.0091	4.3
Cl	15	0.017	2.2	0.21	2.3
Co	16	0.00092	3.0	0.011	2.7
Cr	16	0.00095	4.7	0.012	4.2
Cs	16	0.02	1.7	0.25	1.8
Cu	16	0.0073	4.1	0.09	3.7
Dy	11	0.00042	4.3	0.0049	3.5
Er	5	0.00056	5.4	0.0062	4.1
Eu	7	0.00041	3.0	0.0052	2.4
Fe	16	0.019	2.3	0.23	2.0
Ga	16	0.0024	2.7	0.029	2.5
Ge	4	0.022	1.9	0.3	1.7
Ho	13	0.00042	3.6	0.005	3.0
In	10	0.0079	2.6	0.1	2.4
K	16	0.024	1.6	0.3	1.8
La	13	0.00087	4.8	0.01	4.6
Li	14	0.0022	3.3	0.027	3.1
Lu	1	0.0013	---	0.021	---
Mg	16	0.009	1.6	0.11	1.8
Mn	16	0.00031	4.1	0.0037	3.4
Mo	16	0.00091	2.8	0.011	2.7
Na	16	0.075	5.2	0.91	5.2
Nb	1	0.0058	---	0.046	---
Nd	8	0.0036	2.6	0.0079	4.4
Pb	3	0.01	4.5	0.11	3.4
Pr	1	0.0062	---	0.049	---
Rb	16	0.028	1.8	0.34	2.0
Re	5	0.0011	4.0	0.013	4.0
Sb	8	0.0059	1.8	0.066	1.7
Sc	3	0.0011	2.0	0.012	1.6
Se	3	0.033	2.1	0.43	2.1
Sm	11	0.00066	4.5	0.0076	3.6
Sn	6	0.017	1.4	0.23	1.4
Sr	16	0.00016	2.8	0.002	2.8
Ta	7	0.0035	3.9	0.043	3.2
Tb	16	0.0015	2.0	0.018	1.7
Te	2	0.0084	2.0	0.12	2.3
Ti	6	0.00055	5.2	0.006	4.2
Tl	8	0.0036	1.6	0.04	1.7
U	2	0.00041	1.6	0.0047	2.6
V	1	0.0014	---	0.018	---
W	2	0.007	1.0	0.056	1.0
Y	16	0.00035	3.7	0.0042	3.2
Zn	16	0.12	2.0	1.4	1.8
Zr	8	0.0029	5.6	0.034	4.8

Table 20. Feed-to-meat transfer factors for pig, F^{pig}, (d kg^{-1}) and muscle/feed concentration ratios (CR) sampled from 6 animals. Where N<6, the element was not detectable in all of the 6 samples.

Element	N	GM of F^{pig}	GSD of F^{pig}	GM of CR	GSD of CR
I	6	0.0040	7.8	0.014	7.2
Ag	6	0.021	2.0	0.072	1.8
As	5	0.025	5.8	0.089	5.0
B	4	0.010	3.1	0.036	2.8
Be	3	0.004	6.1	0.015	5.3
Bi	2	0.086	1.2	0.35	1.7
Ca	6	0.0011	16	0.0040	17
Cd	6	0.0045	1.7	0.016	1.6
Ce	2	0.0030	1.5	0.012	1.1
Cl	6	0.028	1.3	0.10	1.5
Co	6	0.0011	2.7	0.0037	2.5
Cr	6	0.0028	2.1	0.0099	2.0
Cs	6	0.29	1.3	1.0	1.3
Cu	6	0.0025	2.0	0.0087	1.8
Dy	4	0.0010	5.4	0.0034	5.3
Er	2	0.0018	2.0	0.0059	2.0
Eu	2	0.0022	1.8	0.0072	1.8
Fe	6	0.0017	9.7	0.0058	9.9
Ga	6	0.0031	1.9	0.011	1.7
Ge	3	0.11	1.5	0.36	1.6
Ho	4	0.00030	9.9	0.0011	8.6
In	3	0.019	1.5	0.064	1.5
K	6	0.044	18	0.15	19
La	2	0.0043	1.7	0.018	1.3
Li	3	0.0056	1.6	0.018	1.6
Lu	1	0.037	---	0.12	---
Mg	6	0.013	12	0.045	12
Mn	6	0.00027	2.5	0.00096	2.2
Mo	6	0.0019	1.3	0.0066	1.1
Na	6	0.019	12	0.068	13
Nb	2	0.0015	2.0	0.0049	2.0
Ni	1	0.25	---	0.80	---
Pb	2	0.021	2.6	0.068	2.6
Rb	6	0.26	1.3	0.90	1.2
Re	2	0.0035	1.1	0.011	1.1
Sb	6	0.0065	3.4	0.023	3.4
Sc	1	0.0012	---	0.0062	---
Se	5	0.056	18	0.20	19
Sm	3	0.0013	6.4	0.0049	5.0
Sr	6	0.0011	1.8	0.0038	1.9
Ta	2	0.023	15	0.075	15
Tb	6	0.0029	4.9	0.010	4.4
Te	1	0.049	---	0.16	---
Ti	3	0.00097	1.5	0.0037	1.2
Tl	6	0.015	1.6	0.054	1.6
U	1	0.015	---	0.048	---
W	1	0.038	---	0.19	---
Y	5	0.00034	9.4	0.0012	8.2
Zn	6	0.0099	10	0.035	10
Zr	4	0.0088	2.5	0.029	2.5

Table 21. Livestock feed-to-meat transfer factors (F, d kg^{-1}) for selected elements computed as fresh weight tissue / dry weight feed concentration ratio, divided by estimated ingestion rate.

Element	Beef	Pig	Lamb	Rabbit	Element	Beef	Pig	Lamb	Rabbit
I	0.0018	0.0040	0.0041	0.16	Mo	0.00092	0.0019	0.0077	0.07
As	0.011	0.025	0.027	0.88	Na	0.075	0.019	0.36	2.4
Ca	0.0011	0.0011	0.0047	0.076	Nb	0.0058	0.0015		
Cd	0.0025	0.0045	0.011	0.13	Nd	0.0036			
Ce	0.00078	0.0030	0.000898	0.023	Ni		0.25		
Cl	0.017	0.028	0.043	1.2	Pb	0.01	0.021		0.11
Co	0.00092	0.0011	0.0023	0.27	Rb	0.028	0.26	0.053	5.7
Cr	0.00095	0.0028	0.000315	0.27	Sb	0.0059	0.0065	0.024	
Cs	0.020	0.29	0.06	3.4	Se	0.033	0.056		10
Cu	0.0073	0.0025	0.065	0.32	Sr	0.00016	0.0011	0.0011	0.18
Fe	0.019	0.0017	0.017	0.19	Th				
K	0.024	0.044	0.10	3.7	Tl	0.0036	0.015	0.012	0.70
La	0.00087	0.0043	0.0015	0.033	U	0.00042	0.015		0.0053
Mg	0.0090	0.013	0.052	1.0	V				
Mn	0.00031	0.00027	0.00013	0.006	Zn	0.12	0.0099	0.22	1.0

Table 22. Livestock feed-to-meat transfer concentration ratio (CR) for selected elements computed as fresh weight tissue / dry weight feed concentration ratio.

Element	Beef	Pig	Lamb	Rabbit	Element	Beef	Pig	Lamb	Rabbit
I	0.022	0.014	0.0097	0.011	Mo	0.011	0.0066	0.018	0.0049
As	0.14	0.089	0.065	0.061	Na	0.91	0.068	0.86	0.16
Ca	0.013	0.004	0.011	0.0053	Nb	0.046	0.0049		
Cd	0.031	0.016	0.026	0.0089	Nd	0.0079			
Ce	0.0091	0.012	0.0021	0.0016	Ni		0.80		
Cl	0.21	0.10	0.10	0.080	Pb	0.11	0.068		0.0075
Co	0.011	0.0037	0.0054	0.018	Rb	0.34	0.90	0.12	0.40
Cr	0.012	0.0099	0.000742	0.019	Sb	0.066	0.023	0.057	
Cs	0.25	1.0	0.14	0.23	Se	0.43	0.2		0.72
Cu	0.090	0.0087	0.15	0.022	Sr	0.0020	0.0038	0.0026	0.013
Fe	0.23	0.0058	0.040	0.013	Th				
K	0.30	0.15	0.24	0.26	Tl	0.040	0.054	0.027	0.049
La	0.010	0.018	0.0035	0.0023	U	0.0047	0.048		0.00037
Mg	0.11	0.045	0.12	0.072	V	0.018			
Mn	0.0037	0.00096	0.00031	0.00042	Zn	1.4	0.035	0.52	0.070

3.4 TERRESTRIAL ENVIRONMENT – VEGETATION AND HONEY

3.4.1 Honey, bees and pollen

With only 2 samples, one each from alfalfa and canola fields, there is limited statistical interpretation to apply to the data. Concentrations for I, Cl and U are shown in Table 23.

Table 23. Concentrations of I, Cl and U in crop used by bees (sampled while in bloom), pollen as collected by the bees, bee whole body and honey (note the units for honey are mass per L, not kg). One sampling site for each alfalfa and canola (there were analytical duplicates).

	I (μg kg^{-1})		Cl (mg kg^{-1})		U (μg kg^{-1})	
	alfalfa	canola	alfalfa	canola	alfalfa	canola
plants at bloom	163	16	800	500	5	<1
pollen	42	20	400	900	1	4
bees	3	38	1900	1400	2	4
honey	<3	<3	<100	<100	<0.04	<0.04

Averaging data for alfalfa and canola, pollen concentrations appeared to be notably lower (more than 2.5-fold lower which is approximately the level of statistical significance) than plant concentrations for: Ca, Cd, Co, Cr, K. La, Li, Mg, Mo, Na, Sr and U. Pollen concentrations appeared to be higher than plant concentrations for Pb, which may suggest some dust contamination in the pollen. Pollen and plant concentrations were similar for As, Ce, Cl, Cs, Cu, Fe, I, Mn, Nb, Nd, Ni, Rb, Sb, Se and Zn. There is no apparent unifying reason for these differences.

Similarly, whole bee concentrations appeared to be notably lower (more than 2.5-fold lower which is approximately the level of statistical significance) than plant concentrations for: Ca, Cd, Cs, I, K. La, Li, Mg, Mo, Na and Sr, many of the same elements as for pollen. Whole bee concentrations appeared to be higher than plant concentrations for Cl, Cr, Cu, Fe and Pb. Whole bee and plant concentrations were similar for As, Ce, Co, Mn, Nb, Nd, Ni, Rb, Sb, Se, U and Zn.

Honey concentrations were only detectable for Ce, Co, Cs, Cu, K, La, Mg, Mn, Na, Rb, Sr and Zn, but for these elements honey concentrations were on average 300-fold lower than plant concentrations. More data is needed to assess the generality of this observation to other setting and other elements. The honey is predominantly carbohydrate (sugars), and as a result the elemental composition is very dilute.

3.4.2 Wild Blueberries

Table 24 is the plant/soil concentration ratios (C_r) for blueberries. The agreement with Sheppard et al. (2006) was very good, within twofold. Note that berries often have especially

low C_r values, and the non-detectable values for I show that the C_r for berries was probably lower than the GM value of 0.0039 found among the detectable samples.

Table 24. Berry/soil dry weight concentration ratios (C_r). Blank cells indicate the element was not detectable in any samples.

Element	Number of measured C_r	GM of measured C_r (unitless)	GSD of measured C_r (unitless)	GM C_r including less-than values (unitless)	GM of soil concentration (mg kg^{-1})	GM of berry dry weight concentrations (mg kg^{-1})
I	2[a]	0.0039		0.0015	2.8	0.0092
Ag	0					0.0021
Al	0				16000	
As	8	0.011	4.2	0.011	2.3	0.022
Au	3	0.031	1.0	0.42	0.017	0.00051
B	12	1.5	1.7	1.5	4.3	6.3
Ba	12	0.16	1.5	0.16	74	12
Be	12	0.0016	1.5	0.0016	0.3	0.00048
Bi	4	0.011	3.4	0.0087	0.16	0.0027
Ca	12	0.4	1.9	0.4	3100	1200
Cd	12	0.042	2.4	0.042	0.22	0.0094
Ce	12	0.00037	2.1	0.00037	30	0.011
Co	12	0.0042	1.9	0.0042	6.7	0.028
Cr	12	0.0017	2.2	0.0017	33	0.054
Cs	12	0.032	2.7	0.032	1	0.033
Cu	12	0.12	3.0	0.12	32	4
Dy	12	0.00074	2.0	0.00074	0.84	0.00062
Er	12	0.001	2.0	0.001	0.36	0.00037
Eu	12	0.0074	1.7	0.0074	0.29	0.0021
Fe	12	0.0016	2.4	0.0016	18000	29
Ga	0				5.7	
Gd	12	0.0013	1.8	0.0013	1.3	0.0018
Hf	0					0.002
Ho	12	0.00083	2.0	0.00083	0.15	0.00012
In	8	0.0086	2.5	0.013	0.024	0.0002
K	12	5.9	1.5	5.9	940	5600
La	12	0.00045	2.1	0.00045	14	0.0062
Li	12	0.0015	1.6	0.0015	14	0.021
Lu	0			0.0021	0.1	0.0003
Mg	12	0.17	1.8	0.17	3000	530
Mn	12	0.51	3.0	0.51	230	120
Mo	12	0.12	2.9	0.12	0.65	0.078
Na	11	0.056	2.0	0.052	450	25
Nb	11	0.0032	3.1	0.0038	0.74	0.0024
Nd	12	0.00045	2.1	0.00045	11	0.005
Ni	12	0.038	2.2	0.038	29	1.1
Pb	11	0.0012	2.6	0.0012	23	0.031
Pr	11	0.00045	2.1	0.00045	3	0.0014
Rb	12	1.2	1.9	1.2	13	16
Re	1	3.2		0.21	0.001	0.00094
Sb	12	0.022	4.0	0.022	0.17	0.0037
Sc	11	0.0012	2.1	0.0012	1.9	0.0024
Se	0				0.17	
Sm	12	0.00052	2.0	0.00052	1.9	0.00098
Sn	1	0.15		0.063	0.73	0.21
Sr	12	0.1	2.0	0.1	19	2
Ta	0			0.0035		0.00021
Tb	12	0.00089	1.9	0.00089	0.15	0.00013
Te	1	0.017		0.037	0.041	0.001
Th	0				1.9	
Ti	0					0.65
Tl	4	0.014	1.4	0.0084	0.079	0.0012
Tm	4	0.00084	1.2	0.0007	0.1	8.9E-05
U	1	0.0013		0.0014	0.72	0.001
V	9	0.00052	1.9	0.00052	46	0.027
W	2	0.065		0.049	0.37	0.014

Element	Number of measured C_r	GM of measured C_r (unitless)	GSD of measured C_r (unitless)	GM C_r including less-than values (unitless)	GM of soil concentration (mg kg^{-1})	GM of berry dry weight concentrations (mg kg^{-1})
Y	12	0.001	1.9	0.001	3.7	0.0036
Yb	8	0.0029	1.6	0.0024	0.25	0.00074
Zn	12	0.11	2.0	0.11	57	6.1
Zr	2	0.08		0.045	1.2	0.075

[a] Iodine was below detection limits in 10 berry samples, of these the C_r was <0.002 and in some samples was <0.0001.
Note: soils ranged pH 4.5-6.1, organic matter 3-37%, clay 1-25%.

3.4.3 Crops

The primary intent of the plant sampling was to measure the transfer factors for various plants relevant to the human food chain. A further intent was to obtain a series of plant types at each of several sites, to determine if there was a consistent difference among the plant types. It was not possible to sample the same species at every site, but it was possible to obtain at most sites samples to represent leafy greens (lettuce, spinach, beet greens, kale, onion tops, cabbage, chard, parsley, dill weed, basil), fruits (cucumber, zucchini, watermelon, apple, pepper, tomato), root crops (beet roots, carrot) and seed crops (wheat, barley, canola, corn, pea). The resulting plant/soil C_r values were summarized (Table 25) and interpreted with the aid of analysis of variance. In general, there were statistically significant differences among the plant types (as categorized above). The overall trend is represented by the rank scores in Table 25: these are based on a principle component Eigen values across elements[3] with the highest plant group, leafy greens scaled to 1 and the lowest, seed crops, scaled to zero. These scores show that fruit was in general mid-way between leafy greens and seed crops, and root crops were nearly as low as seed crops. This is confirmed reasonably well by the data for I and Cl at the top of Table 25. In contrast, the data for Se do not show this trend. Throughout the list of elements, there are several other elements that do not support the overall trend among plant types (e.g., U).

The C_r values for garden fruits were very similar to those for wild blueberries (Table 24). Overall, the correlation between log C_r for garden fruits and log C_r for blueberries was $r = 0.90$ ($P < 0.01$), and the median ratio between the two sets of C_r was <2-fold. This suggests that wild and domestic fruits may have similar C_r values.

For many elements, there was a significant effect of site. Presumably these differences result from differences in soil properties and perhaps weather. The soil properties among the sites were somewhat different: the pH varied from 6.3 to 7.8, clay content from 11% to 58% and organic carbon from 2.4% to 8.5%. There is no apparent consistent relationship between C_r and these soil properties.

[3] Only elements measured in all 104 samples were included in the principle component (PC) Eigen values. The first PC is a single quantity computed across elements that is a weighted score of the log C_r values, the weighting factors were derived so that the PC explained the maximum amount of variation among the log C_r values. Thus it represents all the elements collectively (but to varying degrees). The PC values were then transformed so that the PC for the plant type with the highest C_rs (leafy vegetables) was set to 1, and the PC for the plant type with the lowest C_r (seeds) was set to 0. These PC values are used only to illustrate an overall ranking of plant types with respect to C_r.

For most elements, the Site X Plant Type interaction was not statistically significant. Thus, for these elements the effects of plant type were consistent from site to site: if leafy greens were highest at one site, they were highest at all sites. This was not true for a few elements, specifically I and U. It was possible to do more detailed, pair-wise statistical tests with the I results for leafy greens, root crops and seed crops at 8 sites, and these plant types were statistically different (leafy greens>root=seed) at one site only, the same trend was evident but not statistically significant at 5 other of the 8 sites, and the trend was different (and not statistically significant) in 2 of the 8 sites (on these 2 sites C_r values for seed crops tended to be lower than for leafy or root crops). There is no apparent reason for this interaction related to the species sampled, and so the best conclusion is that the effects of plant type varied site to site, and the expected trend was apparently more strongly expressed at some sites than at others.

Soil Kd values for these sites are shown in Table 26. These values generally agree with previous measurements. As noted above, the soil properties were variable but within the range expected of typical garden and field-crop soils. None the less, there were statistically significant correlations between log(Kd) and clay content, pH and organic carbon content. For most elements, when these correlations were significant, they were positive: the Kd values tend to be higher with higher clay content, higher pH and higher organic carbon content.

The correlations between C_r and Kd, across all samples or specific to field crops and soils, were not statistically significant. The overall correlation coefficient across 22 elements was $r = -0.31$: a negative correlation is expected because lower Kd indicates more soluble elements that may then lead to higher C_r.

Table 25. Geometric mean plant/soil concentration ratios (dry weight basis) from multiple crop species collected from 10 sites where field and garden species were found in proximity. The rank score is based on average principle components scores across elements, rescaled so leafy crops=1 and seed crops =0.

Plant type	overall	leafy	fruit	root crop	seed crop
N	≤106	≤19	≤14	≤18	≤26
rank	0.29	1.0	0.45	0.019	0

Element	N	overall	leafy	fruit	root crop	seed crop	GSD
I	103	0.013	0.033	0.006	0.009	0.008	2.9
Ag	103	0.055	0.124	0.070	0.037	0.039	2.8
As	91	0.012	0.007	0.004	0.004	0.027	5.0
B	106	0.57	2.45	1.33	0.75	0.19	4.2
Ba	93	0.049	0.196	0.028	0.075	0.026	3.5
Be	58	0.00083	0.00303	0.00020	0.00075	0.00029	4.0
Bi	33	0.013	0.013	0.029	0.006	0.019	2.0
Ca	106	0.064	0.079	0.133	0.071	0.048	5.6
Cd	106	0.20	0.78	0.21	0.26	0.12	3.2
Ce	97	0.00059	0.00620	0.00021	0.00067	0.00016	5.8
Cl	71	7.9	25	12	7.8	3.2	3.2
Co	106	0.0044	0.0117	0.0077	0.0045	0.0016	2.9
Cr	105	0.007	0.013	0.002	0.003	0.004	5.2
Cs	105	0.0053	0.0219	0.0054	0.0050	0.0028	4.9
Cu	106	0.20	0.27	0.27	0.20	0.20	1.8
Dy	104	0.00048	0.00508	0.00024	0.00068	0.00013	5.1
Er	104	0.00061	0.00617	0.00027	0.00086	0.00018	5.1
Eu	106	0.0027	0.0121	0.0014	0.0025	0.0019	4.3
Fe	106	0.0024	0.0063	0.0019	0.0014	0.0018	2.3
Ga	103	0.00087	0.00768	0.00031	0.00096	0.00031	4.8
Gd	73	0.00078	0.00251	0.00039	0.00080	0.00036	3.0
Ho	106	0.00047	0.0050	0.00024	0.00071	0.00013	5.2
In	98	0.015	0.051	0.028	0.013	0.006	3.6
K	105	0.71	0.10	0.12	1.37	1.81	8.3
La	95	0.00075	0.00683	0.00035	0.00075	0.00021	5.3
Li	96	0.0037	0.0549	0.0054	0.0030	0.0010	7.7
Lu	71	0.0050	0.0151	0.0036	0.0051	0.0024	2.4
Mg	106	0.21	0.73	0.27	0.14	0.19	2.5
Mn	106	0.039	0.101	0.027	0.017	0.045	2.6
Mo	106	1.2	3.8	1.5	0.7	1.0	5.0
Na	84	0.28	3.19	0.22	0.56	0.04	11.2
Nb	96	0.0032	0.0170	0.0015	0.0025	0.0015	3.5
Nd	106	0.00038	0.00429	0.00021	0.00054	0.00010	5.8
Ni	104	0.024	0.018	0.029	0.017	0.017	2.7
Pb	81	0.0024	0.0049	0.0010	0.0020	0.0022	2.5
Pr	85	0.00080	0.00539	0.00032	0.00078	0.00023	4.2
Rb	106	0.25	0.62	0.36	0.25	0.16	3.5
Re	79	0.53	2.56	0.51	0.14	0.30	6.5
Sb	93	0.0074	0.0172	0.0055	0.0049	0.0033	3.5
Sc	81	0.0016	0.0110	0.0004	0.0013	0.0005	4.9
Se	57	1.0	1.0	0.4	0.7	1.6	2.5
Sm	106	0.00042	0.00451	0.00020	0.00064	0.00011	5.1
Sn	31	0.25	0.12	0.97	1.22	0.14	3.7
Sr	106	0.15	1.06	0.18	0.11	0.07	4.4
Tb	106	0.0010	0.0072	0.0005	0.0013	0.0004	4.0

Element	N	overall	leafy	fruit	root crop	seed crop	GSD
Te	25	0.039	0.064	0.050	0.033	0.034	2.2
Th	55	0.0071	0.0136	0.0036	0.0042	0.0090	3.0
Tl	68	0.029	0.116	0.012	0.016	0.055	8.2
Tm	69	0.0013	0.0056	0.0005	0.0012	0.0004	3.2
U	90	0.0019	0.0052	0.0007	0.0032	0.0011	3.1
V	84	0.0019	0.0088	0.0004	0.0020	0.0013	5.3
Y	106	0.00064	0.00547	0.00036	0.00080	0.00021	4.6
Yb	82	0.0019	0.0080	0.0010	0.0020	0.0008	3.0
Zn	106	0.25	0.28	0.23	0.17	0.37	2.1
Zr	106	0.015	0.087	0.012	0.014	0.007	4.5

3.4.4 Corresponding Soil Kd Values

Table 26. Soil solid/liquid partition coefficients, Kd (L kg^{-1}), for soils sampled in association with the plant samples. Where there were statistically significant correlations between log(Kd) and soil properties, the sign of the correlation is shown, otherwise it is blank.

Element	N	GM	GSD	clay	pH	organic C
I	25	0.74	2.3		+	
Al	33	19000	2.4			
As	19	400	1.9	+	+	+
Ba	31	87	1.9	+		
Ca	32	250	1.7	+	+	
Ce	2	5100	1.2			
Cl	26	45	4.1			
Cr	14	4000	2.0	+		
Cu	33	1200	2.1	+		
Fe	21	76000	2.6			
K	31	2200	3.0	+		
Li	21	620	2.5			
Mg	33	460	2.0	+		
Mn	21	25000	2.6			+
Mo	24	68	4.1		+	+
Na	33	12	2.4		-	
Ni	25	2900	1.8	+		
Pb	1	1600	---			
Rb	3	990	8.9			
Sr	33	180	1.8	+		
U	9	90	2.3			
V	24	3200	2.3	+	-	+
Zn	33	250	1.9			-

4. CONCLUSIONS

The assessment of possible environmental impacts of nuclear waste management is especially dependent on information about the migration of elements. This includes migration in abiotic media such as waterways, sediments and soils and migration among biotic components such as food chain transfers. Humans are an end member of both natural food chains based on wild animals and plants, and domestic food chains based on agriculture. In this study, the objective was to quantify transfers of elements to human food products, with emphasis on I and Cl where previous limitations in analytical capability resulted in a scarcity of data. As a result, this report contains hundreds of transfer factors. These can be used as very specific to the food products measured here, or it is possible to generalise the data to more generic classes of food products.

An example of the possibility to generalise, and a notable conclusion from the study, is shown in Figures 4-1 and 4-2. Here muscle/feed concentration ratios for over 30 elements and various groups of animals are plotted versus those for beef. Virtually all the points fall within 2·GSD, the 95th percentile confidence bounds. In other words, the concentration ratios were similar regardless of whether it was a wild ungulate, a wild goose, or a domestic animal. This unifying observation has been noted previously by Howard et al. (2009b), but this study is the first to make the comparison across so many species using a consistent sampling and analysis method.

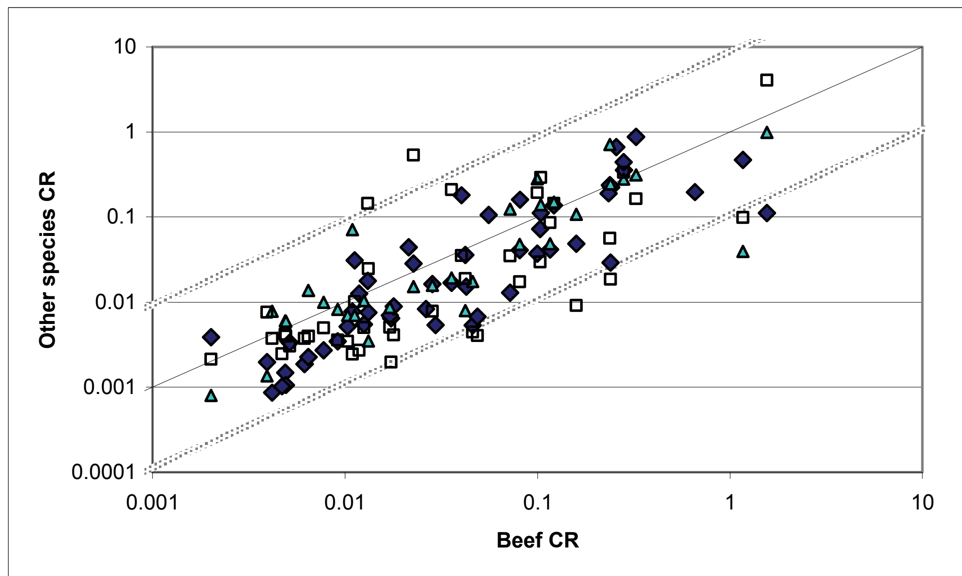

Figure 4-1. Muscle/substrate concentration ratios (CR) measured using stable elements for various species versus those for beef. Each point is an element: the blue diamonds are domestic birds, the turquoise triangles are wild ungulates and the open squares are wild geese. The black line is the 1:1 line, the grey lines are 2·GSD above and below.

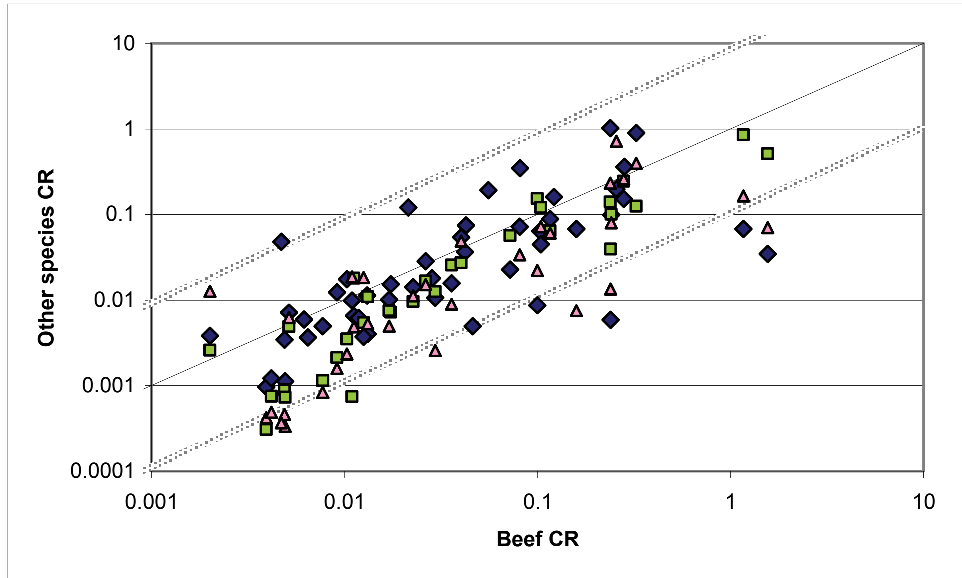

Figure 4-2. Muscle/substrate concentration ratios (CR) measured using stable elements for various domestic meat animals versus those for beef. Each point is an element: the blue diamonds are pig (n = 6), the green squares are lamb (n = 1) and the pink triangles are rabbit (n = 1). The black line is the 1:1 line, the grey lines are 2·GSD above and below.

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APPENDIX A: METHOD DEVELOPMENT AND INITIAL TESTING 2006

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A.1 INTRODUCTION

Although radioactive ^{129}I is the iodine (I) isotope of concern, there is consensus that all isotopes of I will behave in the same manner in the environment. Thus, measurement of stable I in both substrate and recipient media (such as soil and plant, plant and animal) can be used to compute transfer factors to model the potential transfer of ^{129}I among those media (e.g., from soil to plant, plant to animal). Until recently, the most sensitive routine method to measure I in environmental samples was by instrumental neutron activation analysis (INAA), giving a detection limit of about 1 mg kg^{-1} (1 ppm) in plants. This is not sufficiently sensitive to measure I in environmental samples except in materials such as seaweed or in plants grown near the ocean. Accelerator mass spectroscopy (AMS) was more sensitive, but highly specialised and rarely available (the Canadian facility closed in the 1990's). Normal thermal ashing of samples, often used to improve detection limits, cannot be used for I because it is volatile. Thus, measurement of transfer factors was generally limited to experiments where radioactive isotopes could be used, or where stable I levels were elevated above normal. These methods are both artificial and expensive.

Several recent advances have been made in the measurement of I. Inductively coupled plasma mass spectroscopy (ICP-MS) has a good detection level for I in solution, of about 1 mg L^{-1} (1 ppm). Various methods have been used to dissolve or extract I from solids so that ICP-MS or other liquid-based methods can be used for measurement. These include dry ashing with capture of the off-gas, dry ashing in Parr bomb (sealed container), dry ashing with alkaline additives, and alkaline extraction such as with tetramethylammonium hydroxide. Detection levels as low as 2 µg kg^{-1} (2 ppb) in solids have been reported. These detection limits would allow measurement of I transfer factors in many, if not all, plant and animal tissues.

The intent of this project was to implement and investigate one or more of the new methods for dissolution and measurement of I with the intent to identify a reliable, price-worthy method for measurement of I in large numbers of environmental samples at indigenous I concentrations. The media were water and soil samples and plant, fish and animal tissues. A reference material (such as dulce - edible seaweed from Atlantic Canada) was to be identified for QA/QC use.

A.2 SEARCH FOR ANALYTICAL METHOD

The literature was limited to recent studies that report methods sensitive to I at environmental concentrations. The intent was to identify practical methods, Candidate methods were selected (Fecher et al. 1998; German standard methods for iodine (DIN EN ISO 17294-2: 2005); Haldiman et al. 2005; Hejtmankova et al. 2005; Schone et al. 2006; US Center for Disease Control and USDA methods (Caldwell personal communication). On review of these methods, the extraction described by Fecher et al. (1998) and used in the German Standard reported good detection levels and was the least demanding of specialized equipment. It is within the scope of commercial laboratories, and Dr. Eric Hoffman of Actlabs in Ancaster ON became a willing collaborator and expressed interest in working with the alkaline extraction method followed by ICP-MS. There are two disadvantages of this method, both related to the fact that it uses an extraction as opposed to a total digestion. Firstly, extraction efficiency has to be determined for each sample, and at least initially this is done with 3 internal spikes for each sample. Thus each I datum is the result of 4 ICP-MS analyses, which quadruples the cost but does provide good assurance of accuracy. Secondly, although it was envisioned that

multiple element analyses could be obtained from each ICP-MS sample, because this is an extraction proven only for I, the other analytes cannot be reliably measured in the same extract solution. However, routine analysis by ICP-MS is economical, and could be applied to the same samples, thus providing analyses of ~50 elements for the same sampling and sample preparation cost.

The development of the method was done interactively between Actlabs and ECOMatters staff. The internal spike protocol meant that spiked samples as originally envisioned would have little added value, and so these were not done. Samples consisting of various blend ratios of vegetation of known high and low concentrations were prepared as planned, but physical differences between the two pure vegetation samples (ground dulce and ground potato) meant that the blends were physically heterogeneous. When ground together, the dulce, which would not grind to a fine powder, immediately settled to the top of the mixture, due to its larger size. Actlabs staff felt that discrepancies between expected and observed concentrations (if they occurred) could not be unambiguously attributed to non-linearity of the extraction efficiency.

A.3 SAMPLE IDENTIFICATION AND COLLECTION

In order to test the applicability of the method for measuring transfer factors, several types of samples were gathered. There were several objectives. One was to establish the effective concentration range over which the method was useful. Especially important was to establish the lower detection limit. Secondly, it was important to determine that the method gave results that increased linearly with expected concentrations. Finally, it was useful to obtain I concentrations for a range of sample types, including sample origins, in order to be able to plan an effective sampling campaign in future.

There are few commercial reference biological materials for I analyses. Anticipating difficulty, it was decided to develop an operational reference material with consistently detectable I concentration. Dulce (edible seaweed from Atlantic Canada) was chosen, and proved to have about 100 mg I kg^{-1}. Repeated measurements using both the INAA and the alkaline-extraction ICP-MS method were done to provide assurance of the concentration.

Plant samples were expected to have the lowest concentrations and have the most relative variability among the various environmental media. Samples were collected from Nova Scotia, where the oceanic influence should ensure relatively high I concentrations, and from Manitoba where the mid-continent effect should result in very low I concentrations. The types of plants included vegetables, root crops, grains and forage crops.

Animal samples were expected to be less variable in I concentration, because I is an essential element for animals. However, the bio-active I is in the thyroid, and the thyroid itself was not sampled because it is not included in many human foods. Agronomic tissues (egg and chicken, pork and beef muscle) and freshwater fish tissues (goldeye and pickerel muscle) were included.

Soil samples can probably be analyzed successfully by INAA. However, to ensure the data are comparable to the plant results, and in case some soils have very low I concentrations, selected soil samples were also subjected to the alkaline-extraction, ICP-MS method.

A.3.1 Sample type and source

Dulce, commercial, obtained in grocery stores near Halifax
Green bean pods, market garden near Halifax
Green bean pods, private garden in Pinawa, Manitoba
Cabbage head (excluding outer leaves), market garden near Halifax
Cabbage head (outer leaves), market garden near Halifax
Cabbage head (excluding outer leaves), Manitoba-grown
Potato (inner flesh), market garden near Halifax
Potato (skins), market garden near Halifax
Potato (inner flesh), Manitoba-grown
Potato (skins), Manitoba-grown
Barley (grain), Harbourville, NS
Barley (chaff), Harbourville, NS
Timothy, Truro, NS
Egg white, from free-range chickens in NS
Egg yolk, from free-range chickens in NS
Beef (sirloin steak), commercial, from AB
Chicken (breast), commercial, Manitoba-grown
Pork (tenderloin), from Saskatoon, SK
Fish, pickerel, commercial from Lake Winnipeg, MB
Fish, goldeye, from Winnipeg River, Pinawa, MB
Soil, Harbourville, NS

A.4 SAMPLE PREPARATION

Dulce clumps were separated to facilitate drying. Cabbage outer leaves were removed and prepared as a separate sample. The cabbage core was removed, and the rest of the cabbage was cut into strips and pulled apart to facilitate drying. Potatoes were peeled and the peel was prepared as a separate sample. The inner potato was grated to facilitate drying. Barley samples was separated by hand into grain and straw. Timothy hay was separated to ease drying.

Eggs were separated into egg white and yolk. These samples were sent to Actlabs for freeze-drying prior to analysis. Meats were trimmed of fat and thinly sliced to facilitate drying. Fish muscle was removed from bones and skin and thinly sliced to facilitate drying.

The samples were laid on non-stick aluminum foil, weighed and placed on trays in a Nesco American Harvest Snackmaster Dehydrator. Temperature did not exceed 35°C. Samples were checked periodically, then removed when dry, weighed and placed in plastic bags. The samples were then ground using a Braun Coffee Grinder KSM 2B, and placed in plastic bags.

A.5 ANALYTICAL METHOD DEVELOPMENT FOR IODINE ANALYSIS

A.5.1 Iodine extraction procedure

Samples were accurately weighed into Teflon digestion bombs (~0.5 g). Five mL of deionized water were added to each bomb and the contents were mixed to obtain a homogeneous suspension. One mL of 25% tetramethylammonium hydroxide (TMAH) was added to each

bomb and the bomb was closed immediately. The bombs were placed in a water bath, preheated at 90°C for three hours. After heating, the bombs were allowed to cool to room temperature and were opened. The final volume was diluted to 25.0 mL with deionized water. The sample solutions were quantitatively transferred to centrifuge tubes and centrifuged at 5000 rpm to remove the residue. Finally, a portion of the solution was filtered through a 0.45 µm membrane syringe filter. For high concentration samples, additional dilution was sometimes required.

A.5.2 HR-ICPMS Analysis

The method detection limit for I determination by HR ICP MS was 2.8 µg L^{-1} (ppb), computed as the average of the sample blanks plus 3 standard deviations of the average of the blanks. It was concluded that the quantitative analysis of I must be performed by using standard addition calibration to avoid matrix effects. A blank, one unspiked and two spiked aliquots must be analyzed for each sample.

A.6 RESULTS

A.6.1 Results for Reference Standard Materials

Three NIST certified reference materials covering a range from 0.006 µg g^{-1} to 0.855 µg g^{-1} of I were used to evaluate the method extraction efficiency. The variation on the certified value for the durum wheat sample is rather high, and so it is not the preferred commercial reference material. The results are as follows:

NIST RM number	Description	Certified value (mg I kg^{-1} dry wt)	Measured value (mg I kg^{-1} dry wt)
NIST RM 8436	durum wheat grain	0.006 ± 0.004	0.004
			0.003
			0.006
			0.005
NIST RM 8433	corn bran	0.026 ± 0.006	0.027
			0.024
NIST RM 1573a	tomato leaves	0.855	0.738
NIST SRM 8414	bovine muscle powder	0.035	0.043
			0.042

The results from the analysis of reference standard materials were promising and repeatable and so analysis for I in the various environmental samples were undertake. Some samples with higher expected I concentrations, were analyzed using both INAA and the akaline extraction ICP-MS methods.

A.6.2 Results of Samples Analysed by Both Methods

Sample Identification	**Alkaline extraction ICP-MS** (mg I kg^{-1} dry wt)	**INAA** (mg I kg^{-1} dry wt)
Dulce	87.8	99.8
Dulce	109	98.2
Egg white	0.288	< 0.5
Egg yolk	2.650	2.4

A.6.3 Results of Blended Samples

Sample Identification	Expected value (mg I kg^{-1} dry wt)	Measured value (mg I kg^{-1} dry wt)
Dulce/potato blend 1/9 rep a	9.9	9.830
Dulce/potato blend 1/9 rep b	9.9	8.260
Dulce/potato blend 1/99 rep a	1.0	1.110
Dulce/potato blend 1/99 rep b	1.0	1.520
Dulce/potato blend 1/49 rep a	2.0	0.890
Dulce/potato blend 1/49 rep b	2.0	1.740

A.6.4 Results of Various Other Environmental Samples

Samples were analyzed in duplicate. Two aliquots of Harbourville soil were analyzed.

Green bean pods, market garden near Halifax	0.068	0.064
Green bean pods, private garden in Pinawa, Manitoba	0.019	0.018
Cabbage head (excluding outer leaves), market garden near Halifax	0.051	0.048
Cabbage head (outer leaves), market garden near Halifax	0.098	0.103
Cabbage head (excluding outer leaves), Manitoba-grown	0.017	0.018
Potato (inner flesh), market garden near Halifax	0.012	0.011
Potato (skins), market garden near Halifax	0.091	0.095
Potato (inner flesh), Manitoba-grown	0.005	0.005
Potato (skins), Manitoba-grown	0.020	0.020
Barley (grain), Harbourville, NS	0.012	0.014
Barley (chaff), Harbourville, NS	0.302	0.281
Timothy, Truro, NS	0.209	0.213
Beef, commercial, from AB	0.175	0.119
Chicken, commercial, Manitoba-grown	0.022	0.018
Pork, from Saskatoon, SK	0.023	0.029
Fish, pickerel, commercial from Lake Winnipeg, MB	0.063	0.066
Fish, goldeye, from Winnipeg River, Pinawa, MB	0.034	0.036
Soil, Harbourville, NS	24.5	19.5
Soil, Harbourville, NS	20.8	21.7

A.7 CONCLUSIONS AND RECOMMENDATIONS

The results with the new analysis method showed measurable iodine concentrations in all samples tested in a range from 0.005 to ~100 mg kg^{-1}. Reproducibility was good except for the dulce/potato blends, where sample physical heterogeneity (the dulce did not grind well) affected results. Linearity was assessed only for the blends, and here the tenfold concentration range subjected to analysis did give about a tenfold range in results. Costs will be higher than anticipated. One sample analysis costs $100. for ICP-MS and with the blank and 2 internal spikes this raises the per sample cost to $400. However, with experience it may be found that

the numbers of internal spikes can be decreased, to save costs. For example, if a sampling plans call for analysis of 50 fish muscle tissues, this may provide a sufficiently uniform media that only some sample analyses need to include internal spikes.

Overall, the method will be useful to determine transfer factors for I under natural conditions.

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