Isotopic dilution model for ^{36}Cl in the French landscape

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Abstract. Chlorine-36 is one of the most critical radionuclides to consider in intermediate- and high-level nuclear waste management. Special attributes are long half-life, high mobility, biologically essential element, and massive isotopic dilution in the geosphere and biosphere. A model was developed to predict the radiological consequences of ^{36}Cl to human dose receptors. The approach had three parts, a literature and benchmarking review of other ^{36}Cl models and programs, the development of a ^{36}Cl model suitable for ANDRA applications within Aquabios, and the measurement of stable Cl concentrations and parameter values in the relevant landscape. A major issue was to resolve the degree to which specific activity relationships were useful. Soil-to-plant and aquatic food pathways were modeled using specific activity relationships. For subsequent transfers, the model used more traditional transfer factors. The use of the partial specific activity model simplified the problem and made the model and parameters more general. This is especially relevant for transfers of ^{36}Cl because the concentrations of stable Cl in the environment can vary substantially as a result of oceanic effects and local mineralizations. The preliminary field study confirmed some of the key parameters to add confidence to the chosen parameter values and their ranges.

1. INTRODUCTION AND BACKGROUND

Nuclear waste agencies worldwide are working toward the deep geological disposal of high-level radioactive fuel wastes deep in stable rock or clay formations. The radionuclides that are long-lived, most soluble in groundwater, and therefore most likely to leach from these fuel wastes in time are ^{129}I, ^{14}C and ^{99}Tc. Recent analyses of fuels [1] along with the expectation that ^{36}Cl would have release and transport behaviour similar to ^{129}I, has prompted many countries to reassess their ^{36}Cl inventories and add ^{36}Cl to their modelling approaches. Special attributes of ^{36}Cl are that stable Cl is biologically essential, and there is massive isotopic dilution of ^{36}Cl in the geosphere and biosphere. It is nonvolatile (not subject to vapour transport at ordinary temperatures).

2. LITERATURE REVIEW AND BENCHMARKING

The major recommendation arising from the literature is that it is appropriate to use aspects of a Specific Activity (SA) modelling approach in the biosphere. For example, Boursier et al. [2] indicated that a five-fold increase in soil concentration did not cause an increase in plant concentration. This implies the plant Cl concentration is physiologically regulated and would not be modeled well by a concentration ratio. A SA model approach is better able to deal with elements that are abundant and physiologically regulated. In a complete SA model, several pools of isotopic mixing are defined, and these pools are combined in some way to compute exposure. In the context of the local or regional biosphere modeling, there is often insufficient data on the interchange between potential mixing pools to undertake a detailed SA model. A partial SA model is most appropriate and is the approach taken here for Aquabios.

We benchmarked how the 13 major nuclear programs worldwide have undertaken safety assessments with respect to High-Level Waste (HLW) disposal including ^{36}Cl. Benchmarking served as a resource, a foundation to model development and eventual parameterisation. Our benchmarking work showed that those programs that did include ^{36}Cl in their biosphere models:

Article published by EDP Sciences and available at http://www.radioprotection.org or http://dx.doi.org/10.1051/radiopro/2002147

- Employed transfer factor models, except for Canada and the UK where partial Specific Activity models were used,
- Assumed no sorption to soil or sediment media, except for the Canadian model,
- Found the ^{36}Cl dose was delivered primarily through plant uptake, and
- Cited coastal proximity as an important factor in the dose assessment of ^{36}Cl.

Many programs are using the same values and there is little differentiation between the meat types (cattle versus sheep). Few nuclear fuel waste disposal programs have a biosphere research component, some have compiled basic data on transfers of stable chlorine between relevant and important biosphere compartments (Canada and Japan) and only one has carried out experiments in the field with ^{36}Cl (UK). Recent advances in measuring ^{36}Cl in the biosphere and geosphere has the potential to enhance the data for future assessment work. The recent use of ^{36}Cl in aging groundwaters will promote the ability to clearly define and measure flow rates at geosphere-biosphere interfaces and will serve to set background ^{36}Cl values for future safety assessment work within each country.

3. BIOSPHERE MODEL

3.1 Conceptual Model

The model is intended to apply to an inland environmental setting consistent with the present climate and agricultural land use. The surface water body is a river. The dose endpoint is an individual or a critical group that draws the majority of its resources (food, water and fuel) from local sources. The model was intended to allow for some variation from the present climatic conditions, in particular a colder climate similar to present conditions near the Arctic Circle. It is not anticipated that a cold climate will affect the basic model for ^{36}Cl, but there may be changes in parameter values. Some crop types may not be relevant, the proportions of various foods in the diet and the proportion of local diet food items may change, residence time in soils will be longer, and residence time on plant foliage may be longer.

3.2 Features of the behaviour of ^{36}Cl that guide model selection

Features that are important to the definition of the conceptual model for ^{36}Cl include:

- Cl is very mobile in the environment, in some cases even more mobile than water because of anion exclusion.
- Stable Cl is present in the environment in predictable concentrations, with distinct natural and man-made sources.
- Cl is readily absorbed and regulated by organisms.
- In human (and non-human) dose assessment, the soil-to-plant transfer of ^{36}Cl appears most important and central to the highest exposure pathway.
- Plants have the potential to absorb much of the Cl present in the soil, so that mass balance issues could arise in some model formulations.
- Cl concentrations in animal tissues are fairly constant.
- Cl concentrations in plant tissues are high and fairly constant.
- Cl is not volatile to any significant degree within the context of natural processes in the biosphere.
- Anionic Cl is the dominant chemical species.
- ^{36}Cl is a beta emitter so that internal pathways will generally be more important than external exposure pathways.
- Isotopic fractionation in the biosphere can be ignored.

Figure 1 illustrates the transfers built into the model. The figure illustrates transfer from contaminated water to primary environmental media, and also indicates which processes were modelled using explicit specific activity concepts.

4. MODEL RESULTS

Results of the introduction of a single unit activity of ^{36}Cl (1 Bq ^{36}Cl L^{-1} well or river water) illustrate the importance of the major pathways to humans and the calculated dose consequence. For a unit activity of ^{36}Cl in river water, the most important pathways are

- Ingestion of plants and fruit,
- Ingestion of animal products,
- Ingestion of fish and aquatic invertebrates and macrophytes,
- Ingestion of soil,
- External dose due to sediments and shoreline activities,
- External dose due to soil, and
- Air inhalation.

These are almost all internal pathways, as expected, since ^{36}Cl is a beta emitter. Dose due to the inhalation of air is much smaller (200 times) than the ingestion of soil. The ingestion of plants is the major dose contributor and this arises because of the very high stable Cl concentrations in plants.

4.1 Dose Limits

As shown in the Canadian and UK Nirex nuclear waste disposal safety assessments the SA modelling approach makes it possible to place upper bounds on the radiological consequences of exposure to ^{36}Cl, a powerful way to place constraints on the uncertainty in biosphere assessments. Johnson et al. [3] showed that isotopic dilution in the geosphere may afford considerable protection of the biosphere. A dose limit was also suggested for use in the present model.

5. FIELD STUDY

The Bure area in France is a potential candidate site for a deep geological repository for nuclear waste. This field study was designed to give preliminary stable Cl measurements for the key parameter values for Aquabios, ANDRA's biosphere model, for this area. Because plants feature so dominantly in the estimated dose, it was deemed important to collect plants, soils and waters to augment and verify the literature data.

5.1 Sample Collection

Samples were collected in various different sites, both in normal and naturally saline environments:

- Group 1 is in natural saline ecosystems with three locations (Lezey, Marsal, Blanche Eglise). For each location, three sample sets of water, soil and halophytes (salicorne, aster) were collected,
- Group 2 included bryophytes and water collected in rivers running on the same geological substrate as Bure (Oxfordian calcareous) : Ornain, Ornançon, Meuse, Orge, Saulx,
- Group 3 samples were composed of soil and rapeseed from different agricultural fields around Bure: in Villiers le sec, Bois de Greux and Gondrecourt le Château, and
- Group 4 samples were composed of vegetables from gardens around Bure. In each garden, plants such as leeks, carrots and aromatic plants (parsley) were collected, as well as soils.

5.2 Data Interpretation and Discussion

The observed water concentrations of Cl (Table 1) were markedly different between the saline and non-saline sites, as expected. Overall, the saline sites were over 70-fold higher in Cl concentration that the non-saline sites. Because the SA model for the risk assessment for ^{36}Cl requires input of the stable Cl concentration in water, this variation is potentially important. The SA model accounts for the isotopic dilution of ^{36}Cl with stable Cl. There would be substantially more dilution at the saline sites resulting in lower dose estimates, perhaps in the same magnitude, about 70-fold lower. The mean concentration for the non-saline sites is about the same as recommended for the assessment model. The soil concentrations were even more different between the saline and non-saline sites (Table 1). Based on the literature reviewed for the ^{36}Cl project, these values span the expected range.

The concentrations in the aquatic plants are about 40-fold higher at the saline than the non-saline sites. These species are not expected to be food or direct food-chain items. They do illustrate the relationship among environmental compartments. Much of the Cl in aquatic plants will come from the water (as opposed to the soil or sediment). For comparison purposes only, the plant/water concentration ratios were computed (Table 2). They are essentially the same between the saline and non-saline sites. This implies that the plant concentrations are dependent on the water concentrations over a 70-fold range in water concentrations. In the proposed model for ^{36}Cl, a SA approach was used for aquatic biota. Implicit in that model is the assumption that stable Cl and ^{36}Cl will behave in the same way. Thus, these plant/water concentration ratios for stable Cl would also reflect the ratios expected for ^{36}Cl.

Table 1. Concentrations of Cl in water, soil/sediment and plants of saline and non-saline sites.

Sample	Water (mg/L)	Soil (mg/kg DW)	Aquatic plants (mg/kg FW)	Aquatic plants (mg/kg DW)	Plant/ water ratio
Mean of saline sites	1730	708	12400	70900	40
Std Dev of saline sites	243	303	5910	38400	21
Mean of other sites	24	4	300	1350	59
Std Dev of other sites	16	3	96	767	18

Concentrations of Cl in garden plants and grass and corresponding soils are somewhat variable, and this is not related to species. For example, the concentrations in leek span 5-fold and the range among all species was only 7-fold. The overall geometric mean (GM) chlorine concentration in the sampled plants on a dry weight basis was 280 mg/kg with a geometric standard deviation (GSD) of 1.9. The overall GM soil concentration on a dry weight basis was 5.9 (GSD of 6.3). These data are directly applicable to the ^{36}Cl model, because the soil-to-plant transfer was modelled as a specific activity relationship, and for this stable element concentrations are required. These plant values are lower than those previously recommended, but the ranges are generally consistent. Tsukada and Nakamura [4] observed a range in Cl concentrations of 210 to 1400 mg/kg in edible plants. They showed pasture grass much higher at 14000 mg/kg, but it is not clear if these were near the ocean. Sea spray will markedly increase plant Cl concentrations. Chant et al. [5] observed concentrations in the range of 600-1000 mg/kg DW in inland lichens. Often lichens have upper range concentrations because they tend to accumulate atmospherically derived elements. Sheppard et al. [6] reported a geometric mean concentration of 620 mg/kg for a range of agronomic and garden plant species from inland settings. Lower concentrations in the plant implies less isotopic dilution, and therefore is conservative for dose estimation. The recommended values for the model were 600 mg/kg for most plants, and 8600 mg/kg for hay crops.

This study for stable Cl largely confirms the default data used for stable Cl. Advances in accelerator mass spectrometry in the last decade have made the analysis of ^{36}Cl possible in natural soil and groundwater samples, opening the way for field investigations. It is now possible to measure ^{36}Cl down to at least one part ^{36}Cl in 10^{15} parts stable chlorine.

6. SUMMARY

Through literature review and benchmarking with other nuclear agencies, a new biosphere model for assessing the impact of ^{36}Cl was developed. It used specific activity relationships along with transfer factor relationships. Input parameters were derived from the literature, and were supported by a scoping field study in the region of the Bure laboratory.

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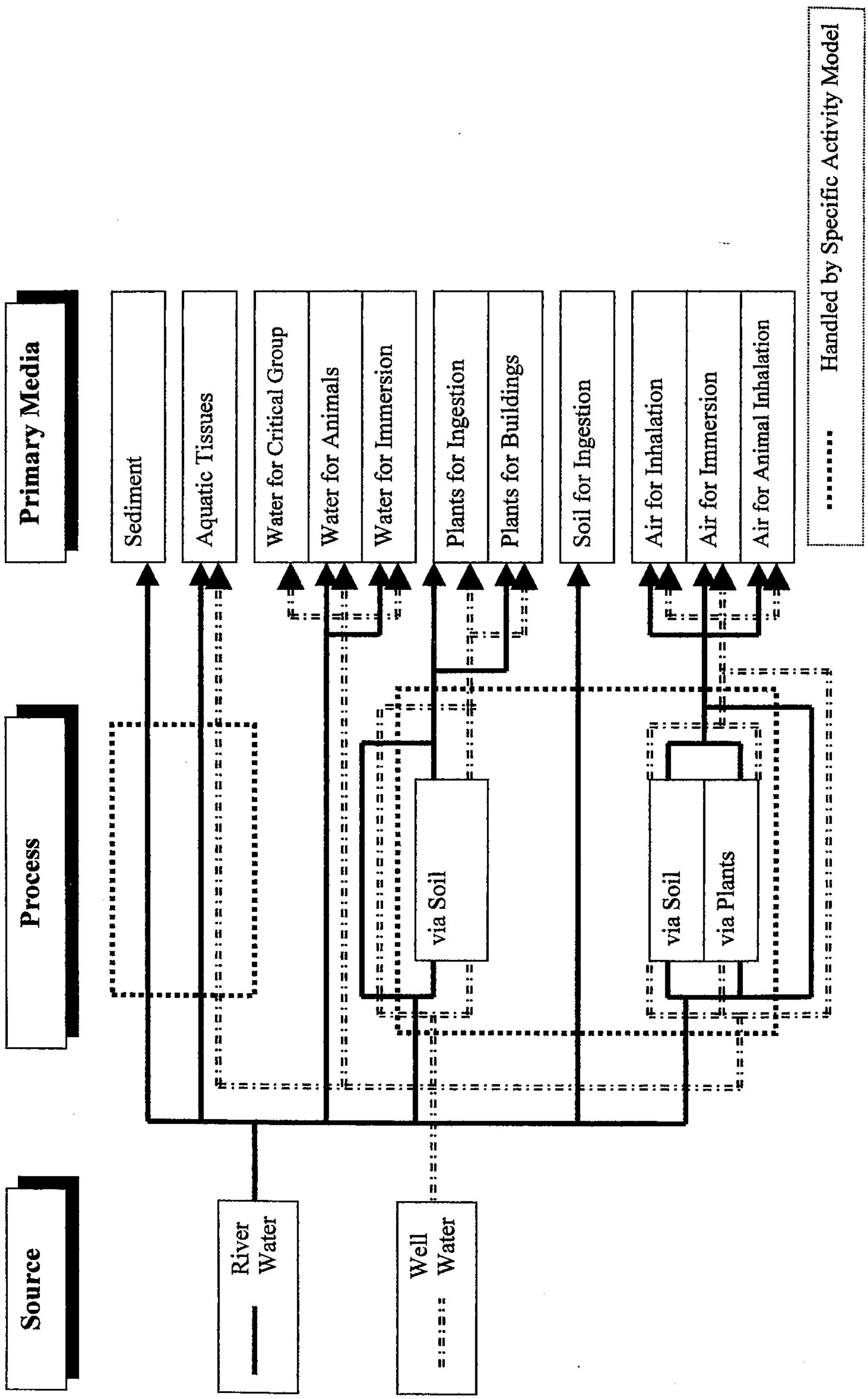

Figure 1 Schematic of partial specific-activity biosphere model for ^{36}Cl