



**Key Issues in Biosphere Aspects of Assessment of the Long-term
Impact of Contaminant Releases Associated with Radioactive
Waste Management**

**Report of an
International Forum on
CI-36 in the Biosphere**

**Châtenay-Malabry, France,
27-28 September 2006**

Hosted by ANDRA

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Preface

The report is presented as working material for information. The content may not be taken to represent the official position of the organisations involved.

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1. INTRODUCTION

CI-36 is an important contributor to potential radiation doses in the long term, arising from release into the biosphere from radioactive waste disposal facilities. Previous work has suggested that the main exposure pathways are ingestion of drinking water, milk and milk products, meat and vegetables and grain products. Special attributes are its long half-life, 301,000 years, high mobility in many environmental conditions, but high uptake into plants, and hence accumulation potential in the food-chain. However, the data necessary for radiological assessment are scarce. This has been demonstrated in various publications but also in the on-going population of the BIOPROTA specialised database.

1.1 Objectives of the International Forum

The objectives of the forum were:

- To provide an open forum for presentation and discussion of environmental processes involved in CI-36 migration and accumulation in the biosphere and on how to model such processes.
- To develop recommendations for the direction of continuing research as input to long-term radiological assessment.

1.2 Acknowledgements

The forum was hosted at ANDRA's office in Chatenay-Malabry, France. Thanks go to Elisabeth Leclerc and ANDRA for organising and providing the venue and hospitality during the forum.

1.3 Participation

There were 29 participants from nine countries, representing a range of operators, regulatory bodies, and technical support organisations. Points of contact are list in Appendix A.

1.4 Report Structure

Section 2 provides summaries of the presentations made and the following discussion. Conclusions and recommendations are given in Section 3.

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2. PRESENTATIONS

2.1 Elisabeth Leclerc: Andra Cl-36 modelling and data requirements

Elisabeth Leclerc presented ANDRA's Cl-36 modelling and data requirements for radioactive waste disposal facilities safety assessments.

The Cl-36 inventories in low and intermediate level waste stores are of interest to ANDRA because Cl-36 contributes 77% ($5.7E-3$ mSv/y) of the peak dose at Centre de l'Aube.

Studies began in 2000 but there are 2 or 3 orders of magnitude uncertainty surrounding Cl-36 transfer and impact without site-specific data. They need to reduce this uncertainty through site characterisations and taking account of site-specific data such as stable chlorine content.

ANDRA is involved in the IAEA EMRAS programme revising TRS No 364. The final draft is due in October and it needs to be populated with reliable data because the report is used by modellers for model input. TRS 364 needs to be updated because critical analysis has highlighted weaknesses and new data have been produced since it was originally published. For each process/transfer parameter there will be associated text to explain the processes involved, co-factors influencing some processes or the determination of parameter values and key equations, so that modellers can make informed decisions on which value to use.

In the current context, the main characteristics of Cl-36 include its speciation in soils. The general assumption is that the predominant chemical species is anionic chloride (Cl^-). However, Oberg (1998)¹ concluded that Cl^- in soils can be converted into various organic forms due to microbial activity and that this clearly impacts upon the biochemical cycling of chlorine.

Chlorine has high mobility in soils which is governed by hydric fluxes (leaching and transpiration). The distribution coefficient between water and soil is very low; Sheppard et al (2006) recommended a geometric mean equal to 0.1 Bq/l.

Chlorine uptake by plants varies by three orders of magnitude in the available literature. Kashparov et al (2005) showed that concentration ratios are not time or soil dependent but the $[Cl-36]/[Cl]$ in the plant does not equal the $[Cl-36]/[Cl]$ in the soil. This relationship is time dependent and so the specific activity approach cannot be used without consideration of the relevant temporal scale issues inherent in any assessment question.

Chlorine is essential to animals; it controls osmotic pressure and internal solution electroneutrality. Recently, Kashparov's work on transfer to cow meat and milk has shown that the isotopic ratio in animal products and foodstuffs is the same. Salt intake does not involve a corresponding increase in the chloride content of muscle tissues and milk.

In radioecology, the transfer factor model, which considers a constant concentration ratio between compartments at equilibrium, is the norm for most radionuclides. Nevertheless, specific activity models are relatively common for H-3 and C-14, because these radionuclides are of elements that are the major constituents of organisms and participate in well-mixed global pools in the environment.

¹ Full references available in the presentation.

The ANDRA models assume no isotopic discrimination, constant stable chlorine contents in environmental media and speciation between chloride and organic chlorine. The main pathway of interest follows from irrigation with contaminated water. A specific activity approach is used which assumes that $[Cl-36]/[Cl]_{water} = [Cl-36]/[Cl]_{plant}$. The main difference between the specific activity and concentration ratio approaches is that in the specific activity approach soil processes are taken into account implicitly. Instead only input from water to different plant compartments and isotopic ratios between animal and water, plant and water and animal and foodstuffs are considered.

The specific activity approach leads to data problems, as the data required for the model are not always available. It is also more applicable to long-term safety assessments than short-term assessments, due to the seasonal variation of chlorine content in soils and the need to take account of all stable chlorine inputs into each compartment in the system including chemical and organic amendment such as fertilisers. However, the specific activity approach does allow chlorine speciation and environmental processes to be taken into account along with adaptation to varying environmental conditions. It can be used to provide a more realistic estimate of Cl-36 concentrations.

Concentration ratio models are used in Sweden, Belgium, Finland, Spain and Switzerland and specific activity models are used in Canada and more recently France. The UK uses both approaches.

ANDRA's future work includes: finalisation of the documents for EMRAS; organising a model intercomparison exercise; launching experimental studies to better understand processes and gain additional data; improving chlorine content measurement techniques; and, long-term evolution of organic chlorine immobilised in soils from decomposition of vegetation.

Discussion

Laurent Rahmani asked if trace quantities of an element in a system would behave the same as large loads and so can be used to populate models. Elisabeth Leclerc responded that we have to assume they behave the same and the same approach is used for all radionuclides.

Kathryn Higley mentioned that the transfer through the environment might not be dictated by the chlorine itself but by the bulk material it is travelling in. The actual chlorine content might not be relevant.

Mike Thorne responded that both specific activity and concentration ratio models look at the degree of dilution of Cl-36 in Cl. The two are not disentangled easily and concentration ratio data are sometimes derived from specific activity models. Specific activity models are better in the long-term but more difficult to use for the short-term. However, this problem occurs due to the way the specific activity models are formulated and by re-writing the formulation in flux terms, but continuing to use the same approach, it could work for short-term assessments.

Frits Van Dorp felt we need to know about long term variability of chlorine in soil rather than concentrating on getting purely site-specific data. For example, fertiliser application is very much connected with present-day behaviour and this may change so we therefore need to know what these activities do to soil to allow future predictions to be made.

Elisabeth Leclerc pointed out that compliance with the requirements of the regulatory body in France means that they only consider present day habits and so do not need to consider how changes in habits may impact a system.

2.2 Kathryn Higley: US perspective on Cl-36 Issues

Kathryn Higley gave 'One person's perspective' on Cl-36 issues because there really is no single US perspective.

Kathryn started with an overview of the origin of Cl-36. She noted that there are both natural and anthropogenic sources of Cl-36. It is naturally generated in the atmosphere by the spallation of argon-36 with cosmic ray protons, and in soil and rock by neutron activation of chlorine-35. The concentration of Cl-36 in groundwater has been used to provide an insight into the age of the groundwater. Large amounts of chlorine-36 were produced by irradiating seawater during nuclear weapons tests conducted between 1952 and 1958. This created what is called bomb pulse Cl-36. On an unrelated note, Cl-36 concentrations in reactor graphite can be used to track the power-level history of a reactor. However to measure Cl-36, some radiochemistry first needs to be undertaken; alternatively, accelerator mass spectrometry is used.

Before discussing Cl-36, Kathryn proceeded to give an overview of the US regulatory framework. This was intended to lend perspective to her analysis of the issue of Cl-36 in the US:

- ◆ US Nuclear Regulatory Commission is concerned with commercial nuclear power and commercial uses (and disposal) of radioactive materials;
- ◆ US DOE is concerned with the production, management, and disposition of weapons material.
- ◆ US EPA considers the performance of legacy and new waste sites. There is some overlap with NRC and DOE in certain areas of regulatory authority.

Kathryn's analysis focused primarily on the US former weapons complex, starting with the Nevada Test Site. The proposed repository for civilian HLW disposal at Yucca Mountain is near the Nevada Test Site. She noted that Yucca Mountain is currently undergoing investigation for a licence application. Key to that application is demonstration of the integrity of the repository structure. In ca 1996, bomb-pulse Cl-36 was detected in test boreholes deep inside Yucca. The implication was that there existed fast flowpaths for groundwater into the site. However, there remains on-going controversy surrounding the analysis. With regard to the license application, the DOE viewed the presence of bomb-pulse Cl-36 as a minor issue that could effectively be addressed through engineering designs (i.e., drip shields for fuel which would limit water intrusion into the waste). With regard to the possible inventory of Cl-36 in the stored waste, Kathryn found no (or limited) discussion of it as a nuclide of concern in future releases. She then discussed Cl-36 at the Idaho National Laboratory. She cited a recent study that suggested for one INL subsurface disposal facility, mobile long lived fission and activation products (Cl-36 being just one of these) posed concern to health under baseline conditions. Most of these products were generated by Idaho Site reactor operations.

She noted that within the context of regulatory oversight, it is often triggered when peak risks are greater than 10^{-6} . The study she cited had predicted risk from Cl-36 in the 100 years period following institutional control rising to somewhere around 6×10^{-6} . She did note that the report emphasized uncertainty surrounding inventory, transport and uptake parameters. Much of the risk is driven by the assumption that the chlorine is completely mobile.

Kathryn went on to talk about the Hanford site. At Hanford discussion of Cl-36 can be found scattered throughout technical reports on the groundwater at the site, and to a lesser extent in documents about the production reactors. Several reports note that Cl-36 is used as a tracer for vadose zone recharge studies. The reports state that its presence in either the vadose zone or groundwater may be due to atmospheric fallout,

a small subsurface production rate, or from reactor operations. However, little information exists on sources or distribution in the groundwater or vadose zone.

A few reports speculated about the presence and fate of Cl-36 released from the fuel processing areas at the Hanford 200 Area plateau. It was expected mainly to stay in the aqueous waste streams and consequently remain with the high activity fission products in the tank waste streams. As to its production, one report suggested that there may be increased concentrations of Cl-36 because chlorine gas was used to remove boron from the graphite moderators in the plutonium production reactors. Ultimately, the fate of this Cl-36 during processing is unknown, but it is considered that a fraction may have been released to the atmosphere with iodine during fuel dissolution.

Kathryn indicated in her review of some Performance Assessments documents for Hanford, she found uncertainty surrounding the Cl-36 inventory. None the less, it emerges as a key mobile radionuclide, with a fraction probably left in place after clean up. The presumption (in the PA documents) is that the source term is overstated and the assessed risks will decrease as more data is obtained.

She then went on to discuss Cl-36 data she found for the Savannah River Site. In reports it was stated that the groundwater is only around 3 feet below the surface of this large site, and the presence of Cl-36 in waters is attributed to both natural sources, bomb pulse, and with a very small contribution from site operations. It is therefore considered that the increase in Cl-36 concentration is just a 'blip' that will pass; irregardless, the concentrations are still well below any regulatory limit.

Kathryn's conclusions from 'grey literature' are that the Cl-36 appears in site waste performance assessments and discussions on its significance continues with site operators, but it hasn't yet reached a level of great concern. This is possibly because sites simply don't carry calculations out long enough. She cited one regulation: DOE order 435.1., performance assessments shall include calculations for up to 1000 yrs after closure. But (she noted) based on observations from Europe peak Cl-36 releases do not occur until around 10000 years after closure. So the U.S. may simply not be looking for it. There also remains the issue of inventory. This isotope may not be accurately modelled in the US codes which estimate source term.

Discussion

Laurent Rahmani asked if any information or data are available on the Cl-36 in disposed graphite and whether or not it was taken into account in any Environmental Impact Assessments. Kathryn Higley did not think it was considered because it has not generally risen to a level of concern.

Charles Meurville mentioned that the Cl-36 in ground water might not only be due to bomb blasts and asked what is the kind of waste which is supposed to be buried around Hanford.

Kathryn replied that the problem can be confounded because the natural cosmic-ray production rate of Cl-36 changes the baseline signature. As for wastes at Hanford, there are lots of different types. Kathryn spoke about the general purpose of Hanford as a plutonium production site in the latter half of the century. She noted that as part of the production process irradiated fuel rods were dissolved and uranium, plutonium, and other valuable radionuclides extracted. The waste streams that were generated during this process were varied and complex. They consisted of solids which were buried in near surface disposal structures; highly radioactive sludges which were stored in million gallon tanks; and millions of gallons of contaminated waste water that were discharged to subsurface structures (tile fields, French drains, trenches) which used the soil as an exchange column to hold the radionuclides. Hanford is no longer used for production purposes, but areas within it are being decommissioned, and

residual wastes removed and repackaged for long-term disposal. In some instances vitrification is being considered.

Pascal Leverd commented that we need to make progress on the Cl-36 inventory data, but asked, 'What is classed as progress?'. This is a general issue for many organisations

2.3 Yves Thiry: The role of forests in the biogeochemical cycling of chlorine: the SCK-CEN approach for a refined biosphere modelling in PA

Yves Thiry presented the possible role of the forest in the biochemical cycling of chlorine. Until now, biosphere modelling has been based on dose calculations following a traditional *a posteriori* approach for a critical group using determined conversion factors for Cl-36 depending on the receptor (well, river, or soil).

Using this approach for a specific case study (repository with a deep water table), the temporal dynamics show that Cl-36 is an important potential contributor to radiation doses. However, no consideration was given to environmental change.

Modelling of the biosphere could be improved by including process-based dynamic modelling, especially in the case of continuous releases. However, climate change also needs to be considered because this will affect the geosphere-biosphere interface zone (GBIZ), biosphere receptors and the transfer rates between biosphere compartments and exposure pathways.

SCK speculate on the future biosphere with necessary assumptions or simplifications; the quantitative descriptions of the functioning and interactions of the current systems emphasises the role of the GBIZ. Prior to speculation on the future biosphere with necessary assumptions or simplifications, priority is given to the quantitative descriptions of the functioning and interactions of the current systems with emphasises the role of the GBIZ.

The nature of the ecosystem is usually affected by the nutrient level and water availability in the soil. SCK have been focussing their studies on the forest ecosystem as a study system for sink-source models, because it is an important part of the landscape everywhere and vegetation development has great effects on water and element cycling. Interactions of climate and site vegetation are well reflected by changes in forest type. Forest areas are often associated with wet zone and riparian areas as a sink for radionuclides. Modern forest process-based models already exist so by using this system there is no need to re-invent the wheel.

There are data gaps in process description and understanding for Cl-36 which need to be understood before it can be simplified for use in models.

During the past 50 years vegetation uptake and adsorption in surface soil layers have been estimated to account for about 25% and 18% of total Cl-36 inventory [Milton et al., 2003]. However there is a lack of quantitative reliable budget, even for stable chlorine. From a hydrological point of view, chlorine is usually considered a conservative element. However, since the 1990's we have known that forest soils can contain and accumulate large amounts of naturally produced organo-halogens.

Biological cycling is a process associated with turnover of organic matter. Forest vegetation is able to retain a relatively large amount of the input to it and thus radionuclides are available in forests for a long time.

Currently SCK is acquiring data and functional information for quantitative descriptions of the radio-element-biomass-water cycling at a forest site affected by a shallow water table to provide ecosystem justification.

The biological cycling model is a mechanistic decomposition of the biological cycle and conceptual modelling in terms of compartments and fluxes; it is driven by annual biomass production.

The site being studied is a pine forest, with the water table 1m to 1.5m deep. SCK are in the process of determining the uptake and cycling of chlorine by vegetation.

There are limitations in the model. It is site-specific with equilibrium assumptions at soil level and no consideration is given to the under-storey and root system. There is also no mechanistic link with radionuclide availability and migration in soil. No causal components (other than a biomass module) are included to make the link with climatic or hydrologic variables.

Importantly the biomass/element cycle cannot be disconnected from the water cycle. The relationship between the chlorine content of rain and the flux of chlorine through litterfall can be used to determine the dry deposition rate.

In conclusion, the forest area (especially in wet zones) is a possible sink and vector of chlorine recycling in the long term due to environmental changes. A description and understanding of forest functioning is a prerequisite to modelling. Data on Cl-36 are quasi absent and stable chlorine cycling is not as well documented as other nutrients, but the biological control of chlorine retention and recycling is obvious. An integrated approach (water-biomass-element) is necessary for a reliable chlorine budget, especially for modelling transfers across the GBIZ.

SCK priorities for Research and Development include: increasing knowledge of chlorine content values for natural vegetation and wild vegetables; direct measurement of uptake fluxes and reliable budgets; quantitative estimates of organic chlorine/inorganic chlorine in plant and soil, which are scarce are required along with the origin and dynamics of chlorine absorbed by a forest zone. Chlorination/mineralization rates in different climatic and ecological conditions need to be determined along with the long-term effect of changes in boundary conditions and the effect of stable chlorine load on Cl-36 recycling.

Discussion

Laurent Rahmani asked if the annual fluxes that SCK have measured could be extrapolated over 10,000 years. Yves Thiry responded that the goal of this project was not to extrapolate these data to another site but to allow increase our understanding of a forest system.

Elisabeth Leclerc mentioned that an agricultural system today could be a forest ecosystem in 10,000 years so a forest in one place today could behave the same as a forest in another place in 10,000 years and so Yves forest knowledge can be applied to other locations.

Laurent Rahmani concern was that although the ecosystem could become a forest, by the time it does, the chlorine will have moved and so no longer be present. Yves responded that it is possible to accumulate Cl-36 in wet systems and so there could be at risk of remobilisation if the system changes.

Philippe Ciffroy said that EdF models generally use mean chloride concentrations in vegetables, yet from Yves presentation it appears translocation through the plant is limited. Is translocation different for different crops and is it necessary to take account of the different chlorine contents in the different parts of the crop? Yves responded

that they have data for concentrations in different parts of the plants and for new plants and old plants.

Mike Thorne mentioned that chlorine is rapidly mobile in the plant in both directions and thus the effect we are seeing is not translocation, but mixing.

George Shaw commented that he found it surprising that there aren't more examples of chlorine flux data in forests because some acid rain programmes did measure chlorine and asked if anyone knew of any examples. Gunilla Oberg replied saying that chlorine measurements do exist but were just used to test hydrology, so the information is held but generally not presented in papers. People need to go back to the original studies. George Shaw is concerned that Cl-36 may not be as good a tracer of water movement as is generally assumed.

Abdelouas Abdesselam wondered if there is a split between organic and inorganic chlorine taken into plants. Yves Thiry replied that the major part of chlorine absorbed by roots is in the inorganic form.

Pascal Leverd stated that Cl-36 is difficult to measure and so how can it be considered a good tracer? Yves replied that it can be relatively easy to measure if you use a mono-specific solution and liquid scintillation counting. This works for high level activity. Mike Thorne added that although Cl-36 can be difficult to measure, it is a good tracer because it compliments C-14 and tritium. Gunilla Oberg also added that chlorine is assumed not react in biological systems so is a good tracer, although its reactivity is under question.

Charles Meurville asked if SCK were able to discriminate between inorganic and organically bound chlorine, to which Yves responded that the values he presented represent total chlorine and were not split.

2.4 Mike Thorne: UK experimental programme and results

Mike Thorne reported some work undertaken with Imperial College (Adrian Butler et al.) on the behaviour of Cl-36 in soils, plants and animals.

During the 1980's lysimeter studies were set up at Imperial College. The lysimeters contained plants growing in soil with the water table fixed at 40 or 65 cm. A buffer tank containing the radionuclides was kept at a set height to keep water table at same height. The lysimeters were based outside so natural environmental processes could continue. The experiments continued for about 6 years in two Phases before moving onto using soil columns. These columns allow control of the water table, including moving it up and down.

With the lysimeters Cl-36 can be added to the bottom where it rapidly redistributes through the soil. Activity balance calculations using a systems model tend to work better when there is some organic partitioning assumed. Soil profiles of Cl-36 develop rapidly, but differently under different moisture stress regimes and the degree of uptake by the plants varies dramatically; from negligible under wet conditions to more than 70% of the total added to soil in dry conditions. Thus, understanding of water flow and water status is fundamental to assessing Cl-36 uptake by plants. There is some evidence for organic partitioning of Cl-36. In chromatographic studies, peaks in concentration of Cl-36 are seen in the humic acid fraction and in a small organic component, and in free chloride form.

Plant-soil transfer factors are usually high but depending on the season they can sometimes be low. These seasonal changes cause the transfer factors to differ by 5 orders of magnitude (0.01 to 10,000). When the transfer factors are very high this is because the plant has removed the Cl-36 from the soil, leaving little in the soil.

There could be a relationship between chlorine uptake and water uptake or it may be between chlorine uptake and the biomass dry rate production. However, both appear correlated and so it can be argued either way.

Mike Thorne presented a simple soil plant model used for assessment purposes. It is a water balance model for soils with a kinetic model of stable chloride flows. It assumes non-linear binding of chloride in soil and both active and passive uptake of chloride by plants. Passive, diffusive loss of chloride from plant roots is included.

Mike Thorne suggested that there were few concerns about assessing Cl-36 transfer to animal products. For inhalation, which is a minor route of exposure, he uses the ICRP Publication 66 model for humans, with compounds of chlorine treated as Class F or Class M for modeling the respiratory system in animals. A four compartment model is used for the gastrointestinal tract, with very little faecal excretion, so as to give rapid and complete uptake of chloride from the stomach(s) and small intestine. Systemic circulation is modelled assuming a well-mixed single compartment. He takes the chloride concentration in milk to be the same as that in the soft tissues.

Mike concluded that:

- Extensive data (at imperial College and elsewhere) exist on transport in soils and uptake by plants in soil columns and lysimeters and very high plant:soil concentration ratios have been observed.
- Binding to organic matter in soil may be significant.
- Uptake by plants is strongly influenced by the water status of the soil and by water flows and/or by biomass production and these vary significantly over short periods of time.
- Detailed mechanistic models of transport in soils and uptake by plants have been developed but model parameterisation is highly constrained by the need to simultaneously fit soil migration and plant uptake.
- Thus a simplified, kinetic assessment model based on the same principles has been developed and both the kinetic soil-plant and animal models can be shown to be consistent with a specific activity approach.

Discussion

Frits Van Dorp wondered whether it was possible with the lysimeter experiments to say that either biomass production rate or water uptake affects chlorine uptake since correlations appear to exist with both. George Shaw responded that the systems are entangled - water uptake leads to biomass growth which is controlled by water availability. Mike Thorne added that we need to include active and passive routes and a backwards diffusive flux to the soil to achieve a self-consistent account of the accumulation of Cl-36 and stable chlorine in plants, as water losses from plants by transpiration contain almost no stable chlorine.

Graham Smith asked if the high transfer factors were taking all of the Cl-36 or all of the chlorine out of the system. Mike Thorne responded that the plants take all the chlorine out of the groundwater that is coming up from depth in the summer because it is dry, but this does not necessarily mean that stable chlorine already present in the soil is necessarily depleted. This depends on the exchange kinetics between chloride in groundwater and that present in the soil. In winter, the plant may get most of its water from recent meteoric water rather than groundwater, so the specific activity drops to nearly zero.

Philippe Ciffroy commented that the concentration ratio concept is valid under equilibrium conditions because in this case equilibrium is reached between soil and water. Mike Thorne added that almost all concentration ratios reported in literature are empirical measures which have never justified if equilibrium exists or not. Equilibrium is not helpful with chlorine, as it is unlikely ever to be achieved. For example, the concentration ratios are high because there is nothing left in soil due to the kinetics of depletion, so all you would need to do is add water containing stable chlorine and the concentration ratio would become low. We therefore need to use a flux based system rather than a ratio based system.

Charles Meurville added that instant equilibrium is usually assumed with tritium models; can equilibrium be achieved with the same speed with chlorine? Mike Thorne responded that he does not use an equilibrium approach with tritium, rather he uses a kinetic specific activity/flux approach. Charles Meurville asked whether the kinetic model is still required for longer-term chronic releases, to which Mike responded that the detail is not required and so data can be averaged. However, care has to be taken with such averaging and a detailed kinetic model is still likely to be required, because with a non-linear model use of the average values of the inputs does not, in general, give the average values of the outputs.

Kathryn Higley pointed out that you would need to be careful with averaging over a year because there are peaks at certain times so if the harvest were to coincide with a peak the dose would be higher. Mike Thorne responded saying that he does the calculation using a kinetic model for a year and averages afterwards to get an annual dose. This is because you want to get an average of dose which is not derived from an average of input parameters.

It was mentioned that some of the Cl-36 was lost during one of the Imperial College experiments and Laurent Rahmani asked if the experiments considered that Cl-36 might be lost to air. George Shaw answered that yes, this was considered and that they tried to measure the loss with traps but have not measured any chlorine volatilisation.

Gunilla Oberg commented that there have been a number of studies looking at volatilisation of organic chlorine but they haven't been able to construct a system that can be connected to chlorine budget yet. Laurent Rahmani and Gunilla Oberg added that they also knew of experiments where Cl-36 had disappeared.

Philippe Ciffroy asked if the speciation of chlorine in soil should be considered for distribution coefficients. Mike Thorne responded that while some sorption can be found in some circumstances, all the K_d s are low and inconsequential. However, he also noted that incorporation in organic matter cannot necessarily be described in terms of k_d values.

2.5 George Shaw: Fate of Cl-36 in the soil-plant system through isotopic exchange processes

George Shaw described a set of experiments undertaken by Stamatia Massoura on behalf of ANDRA. She has undertaken three types of experiment: batch experiments; two soil column experiments which looked at soil migration and plant uptake; and the solid-liquid partition in soil, and related uptake by plant roots. One of the objectives of the experiments was to examine the use of isotopic exchange as a method to understand relative behaviour of stable chlorine and Cl-36. The column experiments were used to follow the dynamics of evolution of soils and plants. The environmental and agricultural conditions which might influence uptake into crops were also examined.

For the batch experiments, four soils were used, 3 collected from within the vicinity of the Meuse site. One was brown acidic soil with an organic matter content of 2.6%, one was a fluvial soil (organic matter content, 3.6%), one was a rendzina (organic matter content, 4.8%) and the fourth was a garden soil with 8.1% organic matter.

The soils were used in batch and the Cl-36 was uniformly mixed. The experiment ran for 110 days and solution samples were taken at various stages (immediately, short and long term). Immediately, less than 100% of the Cl-36 added could be recovered, suggesting that there is rapid exchange of the Cl-36. The garden soil (highest organic matter) showed the largest reduction in the amount of chlorine which is available for extraction. This reached a maximum of 13% irrecoverable, so even in garden soil the majority of the chlorine is still recoverable.

Throughout the period of the experiment, K_d values were low (but not zero) but they were still increasing at the end of the experiment particularly in the case of garden soil which had the highest organic matter content.

Chloride ions were extracted using de-ionised water or NaOH. Higher recovery of chloride is achieved with NaOH and most is recovered from garden soil. This is interpreted to mean that there is some interaction of Cl-36 with the organic matter.

The next experiments involved incubation in soil under controlled conditions. Small containers of soil uniformly mixed with chlorine but held under different conditions (some saturated some not, and at different temperatures, from -20°C to +20°C, and with different levels of fertilisation to alter the stable chlorine content). The evolution of stable chloride in these containers was examined. Over time there was a systematic decline in chloride concentration in soil water. This suggests that the chloride is interacting with the soil matrix. The only case this did not happen for was the soil stored at -20°C. For this case, there was an initial dip in concentration but then it increased, suggesting that there is a positive role for microbial activity at higher temperatures, but that at -20°C the microbes are likely to be inactive.

The column experiments are slightly more complicated. The idea was to follow the dynamics of chlorine and Cl-36 in the soil plant system when two types of contamination (irrigation from above and the water table from below) are present. Two types of soil (colluvial and garden) and two types of plant – *Lolium perenne* (Ryegrass) and *Beta vulgaris* were tested.

For the experiments involving a contaminated water table, the water table was held at a fixed level and was initially uncontaminated. Contamination was added to the bottom of the column and no water was added to the top, meaning that the only water movement was upwards.

For the irrigation system, dosing with contaminated water was delivered continuously from the surface and was free to drain through the soil through a drain in the bottom of the column.

Surface irrigation produced a better yield of crop biomass than water from beneath. Only around 1-2% of the Cl-36 was removed from the system by the plant. However, there tended to be more removal of Cl-36 following irrigation. This is thought to be because the Cl-36 is more or less directly available for root uptake due to the roots being near the surface. The opposite is true for the contaminated water table experiment.

For the contaminated water table experiment, the evolution of Cl-36 in the soil column shows that close to the water table the concentration of chlorine rises rapidly and reaches a steady state, while the top 5 cm of the soil contains hardly any Cl-36.

In conclusion, migration of chlorine appears to be less in the colluvial soil than in garden soil; however due to the organic content of the soils, one might expect it to be the other way round. For stable chloride, there was generally a reduction with time in chloride in soil solution. This is most likely due to removal by growing plants. For the batch experiments there was exchange of chlorine between the liquid and soil phases but the removal from solution was not very dramatic. It could be that the experiments were not conducted over a long enough timescale to reveal the 'real' kinetics.

Discussion

Laurent Rahmani asked if the stable chlorine and Cl-36 were injected to the experiments in the same chemical form. George Shaw confirmed that the stable chlorine that was measured was 'native' chloride. It was separately noted that Cl-36 was injected as a chloride, and it might have behaved differently had it been injected in a different form. Chloride was injected because that is the most mobile form and it has always been assumed not to react, but it can be converted into other forms.

Yves Thiry asked what is the ideal time for an isotopic exchange test. George Shaw replied that the experiments were not conclusive on this point.

Pascal Leverd commented that if the change in behaviour in the -20°C sample was due to the lack of microbial activity, should this change not also have been seen in the sample stored at 0°C. George responded that there was some question over the results but when the sample is frozen (-20°C) other processes are also taking place, for example salts (including Cl⁻) are pushed into solution.

2.6 Christian Tamponnet: Geochemical cycles of Cl-36 in the environment

Christian Tamponnet gave an overview of geochemical cycles of Cl-36 in the environment. Inorganic chlorine compounds are highly soluble and thus do not tend to remain where they fall. They are not major constituents of igneous or metamorphic rocks and are highly mobile in soil, which results in most of the chlorine which lands on land ending up in oceans. However, if it cannot find a path to the ocean it will accumulate on land.

Globally, chlorine is located in a few large reservoirs: the earth's mantle, crust and oceans. The troposphere, freshwaters and pedosphere are the main reservoirs of anthropogenic sources of Cl-36.

Chlorine is found in both gaseous (HCl) and particulate form (mineral aerosol and salt) and wet and dry deposits are similar in magnitude. Chlorine can transfer from the pedosphere to troposphere via two routes, mineral aerosols and biomass burning. Mineral aerosols are injected in troposphere via strong winds during dry season in deserts. Biomass contains chlorine and around 70 to 85% of this chlorine goes to atmosphere during biomass burning. Around 90% is inorganic, the rest is organochlorinated compounds.

The geochemical cycle of chlorine is driven by physical (wind, water, fire) and chemical forces (ion exchange, weathering, rainfall). It has been studied for decades. Therefore, there is extensive information about geochemical cycling of chlorine either in its main inorganic form of chlorides or in some organic forms.

The biogeochemical cycle of chlorine is much more complex and has only been considered quite recently. Indeed, it has been shown recently that organic chlorine compounds not only are the product of human activity but also are of biological origin.

2.7 Gunilla Oberg: The biogeochemistry of chlorine in soil and the global chlorine environmental cycle

Gunilla Oberg presented the biogeochemistry of chlorine in soil and the global chlorine cycle. There is a geochemical cycle of chloride in the environment and it seems there is organic chlorine in different parts of the environment. However, does this organic chlorine influence the geochemical cycle, where does the organic chlorine come from and what is it doing there?

Gunilla stated that it does not matter where in the world you are; organic chlorine will be present in the organic matter in soil. Anthropogenic organo-chlorines are created in bleaching in pulp and paper manufacturing, in the manufacture of pesticides, and in manufacturing of flame-retardants, cooling media, and solvents.

The terrestrial chlorine cycle in the Stubbetorp catchment has been studied. In the top metre of soil storage there is about double the quantity of organic chlorine to inorganic chlorine (18 to 7 g m⁻²). In vegetation there is an order of magnitude more inorganic chlorine compared to organic (2.1 to 0.1 g m⁻²). The inorganic chlorine arises from rain and is 0.4 g m⁻² y⁻¹. The infiltration rate is 0.7 g m⁻² y⁻¹, therefore dry deposition rate is about 0.3 g m⁻² y⁻¹.

Organic chlorine in rain is < 0.01 g m⁻² y⁻¹, and infiltration is 0.2 g m⁻² y⁻¹. Therefore the dry deposition rate is 0.04 g m⁻² y⁻¹. 20-50% of the chlorine coming out of top layer of the soil is organically bound, so there must be a conversion from inorganic to organic chlorine in or on plants. Based on information from different studies it seems the conversion from inorganic to organic chlorine is about 0.1 - 0.3 g m⁻² y⁻¹.

High concentrations of organic chlorine are found in the top soil layers; this decreases with depth, whereas the Cl:C ratio increases with depth. Throughfall has a high Cl:C ratio as does soil leachate and surface water implying that the mobile organic matter is more chlorinated than the immobile organic matter in litter and top soil. The chlorinated organic matter more easily follows the movement of water.

A suggested theoretical framework for the chlorine cycle in soil is that a considerable part of the chloride brought to the system through deposition is transformed to organic chlorine due to exo-enzymes that enable degradation of organic matter by formation of reactive chlorine. The reactive chlorine oxidizes, depolymerizes and chlorinates the organic matter. As a result, the chlorinated organic matter more easily follows the movement of water but eventually it precipitates at lower levels in the soil and is slowly degraded, thus releasing chloride. A considerable part of chloride in surface water originates from decomposing organic matter, not rain.

Discussion

Laurent Rahmani said that assuming that Cl-36 is leached from the subsoil say 40m deep, the chlorine will remain as inorganic and not be converted to organic chlorine. Gunilla Oberg responded that it will as long as it does not come into contact with anything organic. Carbon must be present for the conversion to happen. Yves Thiry added that the resistance to degradation of certain organic chlorines is very high. Gunilla Oberg agreed that when you mineralise organic matter completely you also mineralise the chlorine.

2.8 Graham Smith, on behalf of D Oughton: Cl-36 at Mayak

Graham Smith gave a brief presentation about measurement of Cl-36 at Mayak on behalf of Deborah Oughton of Department of Plant and Environmental Sciences, Norwegian University of Life Sciences.

Mayak is the site of Russian reprocessing and other historic nuclear activities. In the past there were massive releases from Mayak to Techa River (100 PBq) an accident at Kyshtym in 1957 (800 PBq) and discharges to Lake Karachy (20,000 PBq), all of which are hydrologically connected to the Barents Sea.

Soil and vegetation samples were taken close to the site to test for Cs and Sr to reconstruct doses, provide an explanation for health of local population and to help with remediation decisions. Cl-36 wasn't of particular concern. The main problem with respect to health protection arises due to Cs-137 and Sr-90. However, Cl-36 could be used as a tracer to see where the other radionuclides are going. The Australian National University in Canberra has lots of experience using Cl-36 to monitor groundwater movement using accelerator mass spectrometry. Test measurements were therefore undertaken in 2004 on contaminated groundwater but the sample concentrations were too high for the instruments to measure. Deborah Oughton is currently planning to undertake repeat measurements on the groundwater, river water and vegetation samples.

The future application of these techniques and data at Mayak could provide:

- useful insight into the behaviour of Cl-36 in the environment over the longer term, i.e. decades rather than one season,
- better understanding of potential future migration migration of other radionuclides in the soil and sub-soil in the Mayak region, and
- better understanding of GBIZ processes and radionuclide transfers.

2.9 Graham Smith on behalf of Ales Laciok: Alternative models for Cl-36 in different ecosystems

Ales Laciok is responsible at NRI for Performance Assessments for radioactive waste disposal. NRI is currently undertaking dose assessments for those radionuclides which enter the biosphere. Four situations are under consideration: release to farmland, a well; highlands and a small lake. Each situation has different ecology, irrigation and foodchain pathways. The important pathways for radiation exposure for Cl-36 are the consumption of milk, meat and vegetables.

The key issue Ales Laciok wanted to bring to the attention of the meeting is that, according to NRI model assumptions, the main loss from soil is uptake to plants and removal by harvesting. This process is more important than percolation. NRI use a very high assumption for root uptake, similar to the data seen in earlier presentations. This is different for different systems though. In a natural environment, crops die and their constituent elements are returned to the ground but in an agricultural system crops are harvested and the contaminants are assumed to be removed with the crop.

Discussion

Mike Thorne questioned how much Cl-36 is assumed recycled back into the system. Graham Smith could not answer. Mike added that they might like to look at the Nirex 97 performance assessment for information.

George Shaw asked if the Czechs have a disposal site. Graham Smith responded that they are currently in the site selection process and Mike Thorne added that they have several sites under consideration and all are above 500m.

Concerning the significance of geology, Graham showed a graph comparing relative levels of assessed risk obtained when three conceptual geosphere and three conceptual biosphere models are combined. The same Cl-36 inventory is assumed to

be released from the near field in each case. The alternative geosphere models had little impact on the assessed level of risk. However, the alternative biosphere models caused large variations, over several orders of magnitude. The main differences between the biosphere models were different interpretations of chlorine behaviour in soils and crops.

Frits Van Dorp commented that with classic models the geosphere has very little effect on the transfer of Cl-36 because it is so mobile. However, with the Nagra system, a specific activity model must consider the chloride content of the host rock and of the aquifers below or above the host rock. If one aquifer has almost a gram of stable chlorine and the other aquifer has one milligram of stable chlorine there would be three orders of magnitude difference in the results before the biosphere is even considered because the higher the salt content per litre of water the lower the specific activity of Cl-36 per litre of water. Higher salt concentrations are often found in deep aquifers so this is where the Cl-36 from waste is diluted in stable chlorine. The specific activity issue of Cl-36 should be considered from source term to receptor.

2.10 Graham Smith: Cl-36 issues in L/ILW disposal in Japan, on behalf of K Nakai and H Yoshida (JGC)

Graham Smith gave a brief presented the Cl-36 issues in L/ILW in Japan on behalf of K Nakai from JGC Corporation. The main sources of Cl-36 in the waste are activated metals and graphite following activation of Cl-35 contained in metal or graphite as an impurity material. Typical operating waste contains around $1.7E+9$ Bq / t-metal and the decommissioning waste around $6.1E9$ Bq/t-metal.

The release of Cl-36 from the activated metal as it corrodes appears to be the cause of the problem of migration of Cl-36 from the disposal facility to biosphere. There is assumed to be only slight sorption to engineered and natural barriers but they are unsure whether there is larger sorption to organic soil.

Cl-36 is the assessed major contributor to dose for disposal of this waste, assumed to arise due to use of river water for irrigation. However, there are uncertainties in the initial chloride content of activated materials, the release rate from activated metals, sorption and concentration factors.

2.11 Philippe Ciffroy: EdF R&D modelling of Cl-36

Philippe Ciffroy presented details of the EdF R and D compartment model OURSON, incorporating a specific activity used to model Cl-36 migration.

OURSON is a probabilistic compartmental model. There is a parameter database and set of equations determined from an interaction matrix. It is also linked to software to allow uncertainty and sensitivity analyses to be undertaken.

For Cl-36 modelling in OURSON, transfers are governed by the stable chlorine concentration in each of the compartments. Chlorine is essential for life so the concentration in each compartment must be greater than the minimum necessary for life but below toxic levels. Thus, the first step was to build a compartment model for stable chlorine to verify that the content of stable chlorine in each part of the system is appropriate for the levels essential for life.

The transfer equations incorporate a term which makes sure the total chlorine content does not exceed toxicity level and only if the concentration of stable chlorine in the plant is higher than minimum level for growth will the transfer take place.

The same philosophy is used for cattle; the required minimum inputs must be satisfied before the specific activity transfers take place, taking into account foodstuffs and water intake. There are two options; the first considers constant stable chlorine in cattle; the second calculates the stable chlorine content taking account of ingested stable chlorine and the half-life of stable chlorine in cattle.

For calculating doses to man, the relevance of diet is verified and the minimum required chlorine input to man is taken into account.

The parameters required for the model include:

- mean stable chlorine concentrations in river water and soil;
- transfer factors of stable Cl to plants;
- toxicity level of stable chlorine for plants;
- minimum level of stable chlorine for plants growth;
- minimum level of stable chlorine for cattle; stable chlorine in cattle or half-life of stable chlorine in cattle;
- stable chlorine in milk; and,
- minimum level of stable chlorine for man.

Philippe saw the limitations of the model being that specific activity models are based on the assumption that the ratio of Cl-36/Cl in a compartment is the same as the ratio in the environment of the compartment of interest but how exactly is that environment to be defined? It is difficult to determine the relevant temporal and spatial averaging for any particular system.

EdF has assumed that the environment for plants is the irrigation water intercepted on leaves, rain and soil, but there is still a problem of how to weight each of these factors relative to each other.

Philippe Ciffroy also questioned the relevance of traditional parameters used in models such as the soil to plant transfer factor (Bq/kg plant per Bq/kg soil) which is a traditional unit in radioecology but is arguably not applicable for chlorine.

Discussion

Elisabeth Leclerc asked why, if a specific activity approach was being used between the plant and water, we couldn't use a specific activity approach between the soil and plant? This would surely avoid any difficulty in data needs by not using a compartment approach.

Philippe noted that concentration ratios have been discussed and based on Bq/kg (plant) per Bq/kg (soil). The question then arises as to whether the databases should be populated with these data because they are readily available or should the databases be populated with data which is good for the models?

Mike Thorne commented that often in reports the underlying information is not given, preventing the reader from being able to interpret the data for use in their model. We need to get back to using source data, with the assumptions explained so it can be interpreted in different ways for different models. This is the thought behind the BIOPROTA database. However, the thought does not create the relevant original papers.

The relevance of observing the minimum and maximum levels of chlorine in extreme scenarios was also questioned. They are sometimes not respected in performance assessments. Mike Thorne added that if a man is eating 30g of chlorine, the half life in the body will be 1/10 of that if he was eating 3g, and then the ICRP dose factors are not applicable. He concluded that if a specific activity approach was being used in the model, it must continue all the way to the final dose calculation. ICRP calculate the whole body dose based on stable chlorine content of reference man and intake of chlorine because the chlorine is distributed throughout the body. However, if you double the intake should halve the half life in the body.

Mike Thorne mentioned that when using a specific activity approach, the different pools of chlorine should be added together. For example, take irrigation water which contributes as direct water on plant, but a fraction is not intercepted and so lands on the soil adding to the concentration in soil. The specific activity approach should consider the chlorine availability in the intercepted water and that in the upper layers of the soil plus that already in the soil. Cl-36 is like tritium; it will move down soil column when its raining and move back up during evaporation.

Frits Van Dorp added that we also need to look at the minimum and maximum levels of chlorine in crops. The maximum value is crop dependent and so important. Higher concentrations of chloride in crops will produce yield depression. This information should be used as a check on what you are modelling. It could be used to rule out certain scenarios because they produce physiologically un-realistic results

Mike Thorne commented that this information could also be used for coastal situations where the vegetation changes to cope with salt content. Frits Van Dorp added that some plants can adapt to take higher chlorine concentrations.

Yves Thiry mentioned that a similar discussion had taken place in the EMRAS meeting. The specific activity model is pertinent for a long-term system, but if you need dynamics in the system you need to know different origins of entry of chlorine in the compartments. Philippe Ciffroy added that equilibrium models are used in the long term but plants do not reach equilibrium; the timescale is zero because the plants are renewed each year. A plant cannot be 10,000 years old. Mike Thorne mentioned that the stable chlorine content is influencing the processes and thus influencing the uptake of Cl-36.

Graham Smith responded that you could end up with big uncertainties over which area the Cl-36 is interacting. We need to know how the chlorine is mixing within the environment of the plant being modelled. The area of the environment would depend on the nature of the GBIZ, so by including all the necessary additional data to model that, the level of uncertainty could be similar to that in the concentration ratio approach.

Gunilla Oberg mentioned the influence of land use/practices and environmental factors on the migration of chlorine and conversion of chlorine to organic chlorine.

Frits Van Dorp added that the biggest and easiest to justify gains for the specific activity approach are in the near field and geosphere. There is limited impact on biosphere. The only additional sources of stable chlorine to the biosphere are KCl fertiliser or salt licks used for cattle. Mike Thorne commented that these remarks are justified because the likely discharges in Switzerland are to saline waters in the geosphere. The situation is different in different countries. If discharges occur to groundwaters low in salinity, little effective dilution will occur in the geosphere.

3. CONCLUSIONS AND RECOMMENDATIONS

During the presentations, several common themes and issues arose in areas where more work could prove valuable. These included:

- ◆ Distinguishing organic and inorganic chlorine and Cl-36 in samples.

Because of the different mobilities and roles in plant growth etc, it is of interest to be able to distinguish inorganic and organic chlorine. Gunilla Oberg noted that the organic matter cannot be removed first because the process will break the organic matter down. Chloride analysis must be done first, followed by organic chlorine. However, from the samples they have taken, it appears that the variability across 1 m² of forest matches the variability across Sweden. The variation is very large due to the heterogeneous nature of the vegetation and uptake in forests. There may be less variation in pastureland.

- ◆ Understanding of the system so as to resolve modelling/data issues.

Potentially, this could be based on stable chlorine studies. However, the relevant studies will depend on the important dynamic issues in chlorine accumulation and loss from soils, and the relationship to plant uptake in different hydrological conditions. In addition, not all ecological systems will be the same – e.g. coastal, non-coastal, forest and farmland. Thus, repository sites of interest and other aspects of the assessment context may influence choices of relevant studies. These other aspects include features of the assumed geosphere-biosphere interface, the requirement or otherwise to take account of environmental and other system change, and the assessment endpoints that have to be calculated, e.g. the definition of the critical group, and the temporal averaging in the assessed dose that is acceptable.

Gunilla Oberg has done some studies to check if organic chlorine can be found in old deposits/carbon and found that it is present but there is no clear pattern in the quantity found. However, as long as there is carbon in the soil there will be organic chlorine, regardless of the age of the soil. George Shaw wondered how relevant old (geological) carbon stores really are, because coal, for instance, takes much longer than 100,000 years to form and so may not contain the Cl-36 we are interested in. George is concerned that experiments are usually conducted over very short timescales and so scaling the results up may be problematic.

The evolution of forest and European agricultural systems is well known but the evolution of mires, deserts, agricultural systems outside of Europe, coastal systems, tropical and subtropical ecosystems are less well known and so should possibly be considered.

- ◆ Integrated understanding of the plant-soil-hydrological interaction and making distinctions between oxidising and reducing zones.

It was noted that the plant – soil – hydrological interaction should include the interaction with soil organic matter, and that changes can occur through the year, so different seasons need to be considered in appropriate sequences. However, there may also be changes from year to year, which can be relevant for longer-term continuous releases.

Benefit could be gained by performing assessments at different temporal and spatial scales and testing when and under what conditions the results can be extrapolated. The testing would need to be conducted under different hydrological and redox conditions to allow the significance of different assumptions to be determined, based on current understanding.

It was noted that although chlorine is very mobile, organic recycling means that the chlorine is released but then taken up by plants again before it has time to migrate. This gives an appearance of mobility and accumulation at the same time.

- It was proposed to have a modelling task group to look at model development and comparison between specific activity, kinetic and equilibrium models.

The model comparisons should allow for the scientific issues associated with Cl-36 behaviour but also include the radiological endpoint, bearing in mind that the dynamical issues will depend on what we mean by the average dose to a critical group.

The objective may not be to build a completely new model. There are currently two types of model, simple equilibrium and sophisticated, e.g. flux based. However, data are missing for some of the equilibrium parameters. The more sophisticated models may be useful to provide parameters for simple models. In addition, there was consensus towards using dynamic models, and deriving those models on complete system descriptions. A comparison of these descriptions would ensure that all potentially important pathways are included.

- It was concluded that a lot of information had been exchanged among participants with different backgrounds and technical contributions to make to waste disposal safety assessment. In addition, much had been learnt over the course of the forum, which has implications for long term Cl-36 dose assessment.

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