# Characteristics and behaviour of $^{14}$C and $^{36}$Cl in the biosphere in the context of radioactive waste management

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**Abstract.** Evaluation of the safety of high-level radioactive waste repositories is a requisite included in radioactive waste management policy. Criteria for the safety of those facilities require protection of the population and the environment over a time scale appropriate to the characteristics of the radionuclides present in the disposed wastes. In order to demonstrate compliance with regulations, a quantitative analysis of the system must be carried on, including the transport of radionuclides from the facility to the biosphere and through, and the assessment of radiation doses from both external and internal exposure. Previous assessments, demonstrate that some radionuclides contribute more to the annual dose to humans. Among these, $^{14}$C and $^{36}$Cl must be considered. Realistic models to simulate transport of radionuclides in the environment are essential. A biosphere model should represent the transport and accumulation of radionuclides in the different media, and include characterization of the pathways through which the contaminants reach the population giving rise to the exposure to radiations. This paper shows general guidelines for the behaviour of $^{14}$C and $^{36}$Cl in the environment and the definition of a conceptual model of radionuclide transport in the biosphere.

## 1. INTRODUCTION

The evaluation of the safety of high-level radioactive wastes repositories is a requisite of the radioactive waste management policy. Criteria for the safety of those facilities [1] require protection of the population and the environment over a time scale appropriate to the characteristics of the disposed wastes. In order to demonstrate compliance with the relevant principles and regulations, there is the requirement for an analysis and quantification of system behavior including the transport of radionuclides from the facility to the biosphere, their transport in the biosphere and the resulting exposure of humans, including the evaluation of radiation doses from both external and internal exposure.

The biosphere model needs to represent the transport and accumulation of radionuclides in the different media, including characterization of the pathways through which the contaminants give rise to exposure of humans. Such modeling needs to take account of the specific characteristics of the radionuclides involved, which have different physical, chemical and biological characteristics and behaviours and could require conceptually different models to predict their transport through, and distribution in, the relevant environmental media.

Among the key radionuclides, that have been shown to contribute most to the doses estimated in high-level radioactive waste disposal performance assessments [2], are $^{14}$C and $^{36}$Cl. The important factors that determine whether a radionuclide will be a key contributor to dose are: its total inventory, half-life, solubility and sorption potential and biosphere flux-to-dose conversion factor. Both are important in gaseous releases and the chlorine is likely to be more important for the groundwater release and transport pathway.

This paper shows general guidelines for the behaviour of $^{14}$C and $^{36}$Cl in the environment and the definition of a conceptual model of radionuclide transport in the biosphere.

## 2. FORMATION AND INVENTORY

The $^{14}$C is in the residues of the nuclear fuel cycle, as a result of the interaction of neutrons with the atoms of $^{17}$O, $^{13}$C and $^{14}$N present in the irradiated components of the nuclear reactor. This radionuclide has been identified as one of the most significant in the safety assessments, for its contribution to the dose, due to its long half-life, the volatility of the compounds that forms (carbon dioxide and methane) and the easiness with which it is assimilated by the biological systems.

The $^{14}$C occurs in a natural way by the interaction of cosmic rays with the gases present in the upper atmosphere. The main reaction that occurs is $^{14}$N (n, p) $^{14}$C. The $^{14}$C presents a half-life of 5,730 years and emits $\beta$ particles with an energy of up to 155 keV.

In nuclear reactors, the $^{14}$C occurs as a result of the absorption of neutrons by the nucleus of $^{14}$N, $^{17}$O and $^{13}$C involved in the activation reactions $^{14}$N(n, p) $^{14}$C, $^{17}$O(n, $\alpha$) $^{14}$C y $^{13}$C(n, $\gamma$) $^{14}$C. These elements are found in many of the components of the reactor as can be, the coolant, the moderator, the structural materials and the fuel. As a result of these reactions, the $^{14}$C is present in a wide variety of radioactive waste such as spent fuel, ion exchange resins, and pipes of the reactor. The activity of $^{14}$C for fuel element is 5,74 $10^9$ Bq according to the inventory of reference [3].

Chlorine is a very reactive element that does not occur uncombined in nature. The most common chlorine compound is sodium chloride, which is present in seawater, salt wells and large salt deposits, often in association with other chlorides. Chlorine is present in the terrestrial environment as both inorganic and organic chloride. The major source of chloride is the atmospheric deposition originated from the sea, whereas the organic chlorine originates from various sources, taking place its formation and mineralisation in the soil [4].

Chlorine occurs in nature as two stable isotopes: $^{35}$Cl and $^{37}$Cl. Of the seven radioactive chlorine isotopes, only one, $^{36}$Cl, is of concern in the context of solid radioactive waste disposal due to its long half-life, 301,000 years. It occurs naturally in the environment, being produced in the atmosphere, due to spallation reactions on $^{40}$Ar by cosmic rays and, in soils and rocks due to neutron activation of $^{35}$Cl. Anthropogenic inputs of $^{36}$Cl to soils arise mainly from the operation of nuclear industrial processes, where $^{36}$Cl is formed by neutron activation of $^{35}$Cl in materials, such as stainless steel and concrete shielding, associated with the cooling water system. Thus $^{36}$Cl will be present both in waste fuel and construction material originating from nuclear installations.

$^{36}$Chlorine is a low-energy $\beta$ emitter. Two are the decay chains of $^{36}$Cl, one to stable $^{36}$Ar ($\beta^+$) and another to stable $^{36}$S ($\beta^-$). In addition, the decay mode to the latter can occur by electron capture. The significance of this radionuclide is dependent on its inventory, which is currently based on rough estimates. According to [3], the reduced inventory of $^{36}$Cl, 50 years after cooling, is $1,965E + 08$ Bq per fuel element.

## 3. BEHAVIOUR IN THE ENVIRONMENT

The primary pathway of radionuclide releases from an undisturbed repository involves the introduction of radionuclides into the underlying aquifer, contaminating wells used for drinking or agricultural irrigation. However humans could also be exposed to radiation as a result of gaseous emissions from the repository, that in the case of $^{14}$C will be in the form of methane or as carbon dioxide. Due to the ease with which gaseous contaminants are distributed in the atmosphere, human exposure would not be limited to the near-field population, but could extend to a global scale. Amongst the potential gaseous contaminants, $^{14}$C is the one with the highest potential for gaseous release and human exposure [5].

The environmental transport pathways of $^{14}$C are different from those considered for others radionuclides. Whereas most radionuclides are present in the environment in solution or sorbed to solids, carbon can also move through the environment in gaseous form. Moreover, stable carbon is an abundant and ubiquitous element in the environment, and variations in stable carbon cycling will influence the behaviour of $^{14}$C. As for other radionuclides, groundwater is the source of $^{14}$C, and the
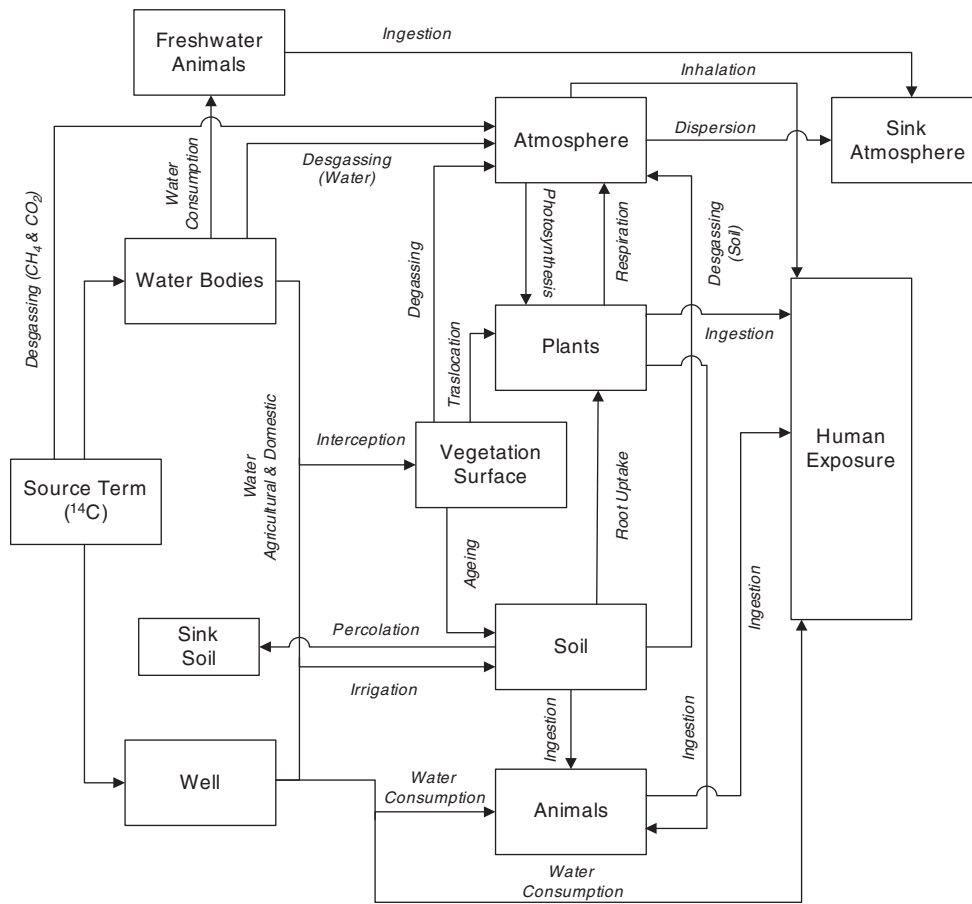
**Figure 1.** Conceptual model of the behaviour of $^{14}$C in terrestrial environments.

calculation of $^{14}$C concentrations in the soil are based on equilibrium relationships between $^{14}$C gains and losses. The most important process resulting in the loss of this radionuclide from surface soil, is the gaseous emission, unique to gases and not considered for other radionuclides. After it is released into the atmosphere, $^{14}$CO$_2$ can be incorporated into crops via photosynthesis.

For the modelization of the behaviour of the $^{14}$C in the different receptors, it is necessary to consider the isotopic exchange with the stable carbon ($^{12}$C and $^{13}$C), the processes in which that exchange takes place and to identify the receptors in which an isotopic balance can be considered. It must also be kept in mind the presence of $^{14}$C in the different compartments, coming from other natural and sources [6].

In the case of $^{14}$C, the primary interactions take place as consequence of processes occurring in the soil zone. Photosynthesis and respiration are fundamental biological processes by which the $^{14}$C moves from the atmosphere as $^{14}$CO$_2$ in the tissues of living organisms.

Soil respiration is the primary path by which CO$_2$ fixed by terrestrial plants returns to the atmosphere. Rising CO$_2$ will increase the flux of CO$_2$ from soils, while simultaneously leaving a greater store of carbon in the soil. Rising temperature and traditional cultivation practices increase the flux of CO$_2$ from soils without increasing soil organic matter. The response of the biosphere to simultaneous changes in all of these factors is unknown, but a large change in the soil carbon pool is unlikely to moderate the rise in atmospheric CO$_2$ during the next century.

Transport of $^{14}$C from an underground burial to a receptor location will be controlled by the movement of groundwater, the amount of water available for transport, and the direction and speed
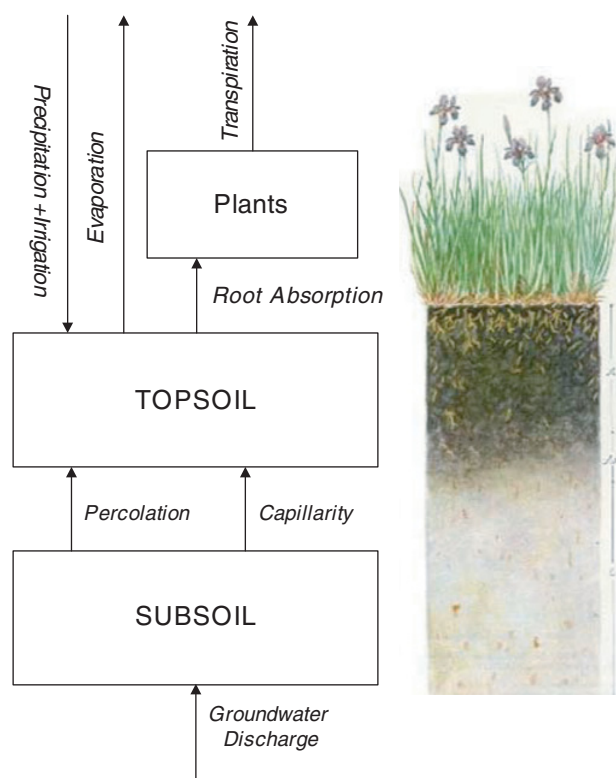
**Figure 2.** Model for the transport of $^{36}$Cl in soils and its uptake by plants.

of groundwater movement. Any physical phenomena affecting the dilution and dispersion of the contaminant will be important in this regard. The transport should also take into consideration geochemical factors, such as precipitation, sorption during the transport, matrix diffusion and isotopic exchange with the more abundant stable C present. Bidirectional gaseous exchange may take place as well. Whereas precipitation depends largely upon other ions in solution and the pH, the mobility of $^{14}$C in the soil-underground is typically represented by the use of a sorption or partition coefficient.

In Figure 1 a conceptual model, that describes the behaviour of $^{14}$C in the biosphere, is shown. This generic model is built following the physical and chemical processes and pathways that the radionuclide takes as migrate through the environment. The model is based on compartmentalizing the biosphere into aquatic and terrestrial components (Atmosphere, Water, Soil, etc.) and then to add the foodchain or animal compartment for humans and animals. A radiation dose is calculated based on quantities for ingestion for internal radiation dose and the timeframes for other activities for external dose.

The two main transport modes for $^{36}$Cl are dissolved in groundwater flow and, in a lower degree, transported in the gas phase. The movement of $^{36}$Cl in the groundwater flow through the geosphere is done through a complex network of interconnected pores and fractures. The transport is due to advection and longitudinal dispersion along fractures and molecular diffusion into the adjacent rock [7]. Depending on the characteristics of the geosphere, several retention processes can contribute to provide a long travel time for radionuclides in groundwater to move towards the biosphere, giving time for the radionuclides to decay and disperse within the system.

In general terms, the geochemical behaviour of $^{36}$Cl, once released into a groundwater system is such that there is no retardation due to the interaction with rocks. It exhibits a high degree of geochemical mobility, moving somewhat faster than groundwater because electrical repulsion from surfaces may prevent it accessing part of the pore space (anion exclusion).

It is expected that any chlorine entering the biosphere from the geosphere will be present as an inorganic species and probably as the chloride ion, $^{36}Cl^-$ [8]. The $Cl^-$ anion does not form complexes readily and, since exchange sites on layer silicates in soil clays are predominantly negative charged, $Cl^-$ tends to be repelled from the mineral surfaces of soil particles. It is often considered not to participate in a significant degree of complexation with organic matter. Chloride shows little adsorption to soil components, being nearly all present in soil solution and is not chemically altered by soil organisms [9].

The solid/liquid distribution coefficient for $^{36}Cl$ in soils is very low [8] and [10], giving rise to its high mobility. This low degree of sorption means that nearly all is present in the soil solution making it available for root uptake and incorporation in plants. According to [8], volatilisation of chlorine occurs, but the impact on soil chlorine concentrations is relatively small.

From the behaviour shown, the most important exposure pathway to $^{36}Cl$ is the ingestion followed in a lower extent by inhalation. The transfer to milk and meat in cattle is high whereas accumulation in aquatic organisms does not occur to any significant extent. Because of weak $\beta$ radiation, the dose coefficients for ingestion and inhalation are low and no significant external exposure.

Figure 2 shows the model for the transport of $^{36}Cl$ in soils and its uptake by plants. The model considers two soil compartments, topsoil and subsoil and two ways of entrance of the contaminant: from the groundwater discharge to the subsoil and in a lower extent due to precipitation and irrigation waters to the topsoil.

## Acknowledgments

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