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Modelling 36Cl in soil-plant system: A phenomenological approach and its application to the discharge of 36Cl in groundwater from radioactive waste deep storage site

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Abstract. Chlorine 36 is considered as part of the group III of radioactive wastes composed of the moderate radiotoxic radionuclides such as Carbon 14. As such and along with its long half-life (3.01×10^5 years) and its chemical form (chloride anion), it is a very important radionuclide to be taken into account in the environment thereby making its modelling an important goal.

We present in this paper the phenomenological approach developed at IRSN for modelling the behaviour of chloride 36 in the continental ecosystems. This model is a part of the SYMBIOSE programme (SYstemic approach for Modelling the fate of chemicals in the BIOSphere and the Ecological systems) whose main goal is to develop a modelling platform able to sustain an environmental chemical risk assessment. Such an approach has been found necessary for Chlorine 36 when the classic models (conventional transfer factor models and specific activity models) fail (for instance in case of acute release of radionuclides). Only the part of the model dealing with the agricultural soil-plant system is described here in detail: interaction matrices, components description, components equations, independent variables, and data.

Then, we present an application of this model to investigate chlorine 36 behaviour in soils and uptake by plants after its discharge in groundwater from a radioactive waste deep storage site. Finally, we present results from an intercomparison study performed under the BIOPROTA international cooperation framework.

1. INTRODUCTION

Chlorine 36 is considered as part of the group III of radioactive wastes composed of the moderate radiotoxic radionuclides such as Carbon 14. As such and along with its long half-life (3.01×10^5 years) and its chemical form (chloride anion), it is a very important radionuclide to be taken into account in the environment thereby making its modelling an important goal [1]. Its ratio to stable Chlorine is about $700.10^{-15}/1$. It comes from the atmospheric spallation of 36Ar and from the neutron capture by 35Cl and muon capture by 40Ca [2, 3]. It has been produced in great quantities during the large scale atmospheric tests of nuclear weapons and is a by-product of electricity producing nuclear reactors and has been found in groundwater [4–8].

2. THE SYMBIOSE MODEL

We present in this paper the phenomenological approach developed at IRSN for modelling the behaviour of chloride 36 in agricultural soil-plant systems. This model has been designed in the framework of the SYMBIOSE programme whose major goal is to improve our capability to predict the fate and transport of radionuclides in ecosystems, and their dose impact on humans, following releases from nuclear facilities under accidental, decommissioning or normal operating conditions [9]. The main challenge

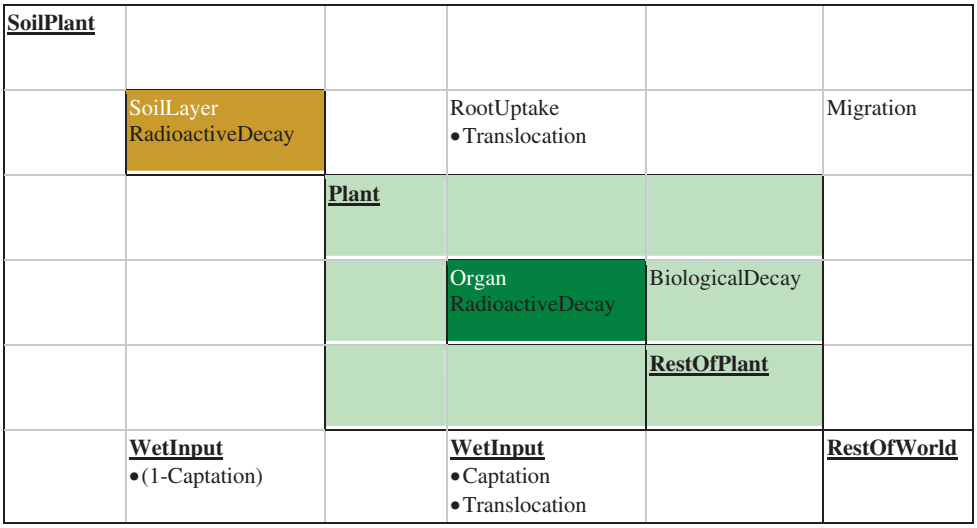

Figure 1. Agri Soil-Plant 36Cl interaction matrix.

was to promote a scientific and software approach that was flexible enough to deal with a wide range of situations, extending from simplified generic studies to more realistic spatially-distributed and site-specific assessments. The SYMBIOSE platform has also been designed to manage data or knowledge issued by research activities in radioecology.

3. THE AGRI-SOIL-PLANT 36CL MODEL

Such an approach has been found necessary for Chlorine 36 when the classic models (conventional transfer factor models and specific activity models) fail (for instance in case of acute release of radionuclides). Only the part of the model dealing with the soil-plant system is examined here, through a description of: components and interactions (basically processes), mathematical approach and data. This model is described in [10, 11]. This model considers that the driving force controlling the movements of stable chloride and Cl-36 is the circulation of water in the system. Contamination may occur via rain (or wet deposition) or irrigation. The case of direct contamination of soil from groundwater was not considered basically in this model.

3.1 Conceptual Model

3.1.1 Components description

SoilPlant (*SP*). Agricultural soil-plant System
SoilLayer (*S*). Part of soil interacting with plants. It is the root layer. Chlorine 36 will only be considered as chloride in soil water. Organochlorines will be neglected.
Plant (*P*). Plants such as grass, vegetables or crops.
Organ (*Org*). Part of plant for human or animal consumption (leaves, grains or fruits).
RestOfPlant (*ROfP*). Other parts of plant not explicitly modelled.

3.1.2 Interactions description

WetInput (*In*). Contamination transfer to the whole soil-plant system via wet aerial pathway: irrigation or aspersion (*Irr*) plus wet deposit (*Rain*).

WetInputPlant = WetInput•Captation (*InP*). Contamination transfer to plant from wet aerial pathway, including interception process by aerial part
WetInputSoil = WetInput•(1-Captation) (*InS*). Contamination transfer to soil from wet aerial pathway, excluding the part of incoming contamination that has been intercepted by plant. Note that WetInput = WetInputSoil + WetInputPlant.
WetInputPlant•Translocation (*InPTra*). Contamination transfer to organ from wet aerial pathway
RootUptake •Translocation (*UpTra*). Contamination transfer to organ via root uptake.
Migration (*Mig*). Contamination migration process from root layer to underlying layers
BiologicalDecay (*Bio*). Organ contamination decay process such as plant transpiration.
RadioactiveDecay (*Rad*). Radioactive decay process.

3.2 Mathematical model

Edible Organ (*Org)*. Evolution of 36Cl concentration in edible organs of plants, in mol kg^{-1} fresh weight, is given by the following equation:

$$\frac{\partial}{\partial t}[^{36}Cl]_{P.Org} = \delta_{Cropping=1}(TCl36_P^{InPTra} + TCl36_P^{UpTra} - TCl36_P^{Bio} - TCl36_{P.Org}^{Rad})$$
$$-\delta_{P=AnnualCrop}(\delta(t - t^{Ger+}) \times [^{36}Cl]_{P.Org})$$

Where the $TCl36$ right hand-side terms (in mol kg^{-1}f.w. s^{-1}) stand for the various processes described above, $\delta_{Croppingg=1}$ is a factor equal to 1 during vegetative period (zero otherwise), and $\delta(t - t^{Ger+})$ is a term used to reset the concentration at the time of harvest, if the plant under consideration is an annual crop.

A similar equation is adopted for stable Cl:

$$\frac{d}{dt}[Cl]_{P.Org} = \delta_{Cropping=1}(TCl_P^{InPTra} + TCl_P^{UpTra} - TCl_P^{Bio})$$
$$-\delta_{P=AnnualCrop}(\delta(t - t^{har-}) \times [Cl]_{P.Org})$$

Adopting physically-based and dynamical parametrizations for each right hand-side terms, and combining equations together, we can demonstrate that this model predicts, in case of a constant wet input flux $TCl36_P^{In}$, an activity in edible organ on the long term that reaches the following equilibrium :

$$[^{36}Cl]_{P.Org}(t \to \infty) = [Cl]_{P.Org}(t \to \infty) \times \frac{TCl36_P^{In}}{TCl_P^{In}}$$

when migration water flux is neglected when compared with root water flux.

Soil Layer (*S*). Evolution of 36Cl concentration in soil layer cultivated with plant P, in mol kg^{-1} dry weight, is given by the following mass balance equation:

$$h_P \times \rho \times \frac{d}{dt}[^{36}Cl]_{P.S} = TCl36_P^{InS} - TCl36_P^{Mig} - \delta_{Cropping=1}(TCl36_P^{Up}) - TCl36_{P.S}^{Rad}$$

Where h_P is the root layer height (m), ρ is the bulk density (kg^{-1} dry weight. m^{-3}), $TCl36$ terms (in mol $m^{-2}.s^{-1}$) stand for the various processes described above, and $\delta_{Cropping=1}$ is a factor equal to 1 during vegetative period (zero otherwise).

A similar equation can be adopted for stable Cl. And here again we can demonstrate that this model predicts a long term activity in the soil layer obeing the following ratio:

$$[^{36}Cl]_{P.S}(t \to \infty) = [Cl]_{P.S}(t \to \infty) \times \frac{TCl36_P^{InS}}{TCl_P^{InS}}$$

When assuming constant input fluxes.

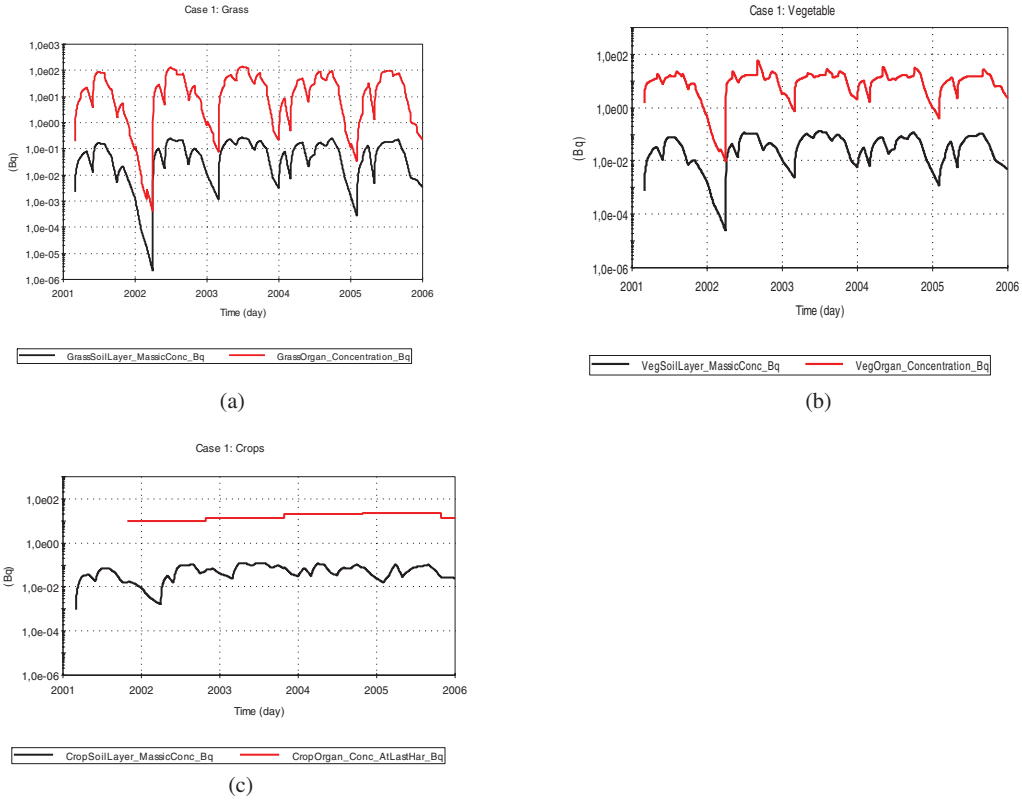

Figure 2. Time evolution of 36 Cl in soil and grass (a), vegetables (b) and cereals (c) expressed in $Bq.kg^{-1}$ (top curves being for plants and bottom curves for soil).

4. APPLICATION TO A SCENARIO OF GROUNDWATER CONTAMINATION

This scenario was given by the BIOPROTA international collaboration forum which seeks to address key uncertainties in the assessment of radiation doses in the long term arising from release of radionuclides as a result of radioactive waste management practises [12].

4.1 Scenario

Then, we used this model to investigate chlorine 36 behaviour in soils and uptake by plants after its discharge in groundwater from a radioactive waste deep storage site. The following scenario was given:

Stable chloride concentration in irrigation water = Stable chloride concentration in groundwater = $30\,mg.l^{-1} = 0.845\,mol\,m^{-3}$. (MCl = 35.5)

36chloride concentration in irrigation water = 36chloride concentration in groundwater = $1\,Bq.l^{-1} = 2.28\ 10^{-8}\,mol\,m^{-3}$. (Specific activity = $4.38\ 10^{10}\,Bq.mol^{-1}$)

There are 2 different locations: inland and coastal sites (BURE & LILLE)

Stable chloride concentration in rain water = $10\,mg.l^{-1} = 0.282\,mol\,m^{-3}$. (MCl = 35.5) in coastal site (LILLE)

Stable chloride concentration in rain water = $2.5\,mg.l^{-1} = 70.4\ 10^{-3}\,mol\,m^{-3}$. (MCl = 35.5) in inland site (BURE)

Rain and evapotranspiration (ET calculated from Turck's equation) fluxes were given for both sites.

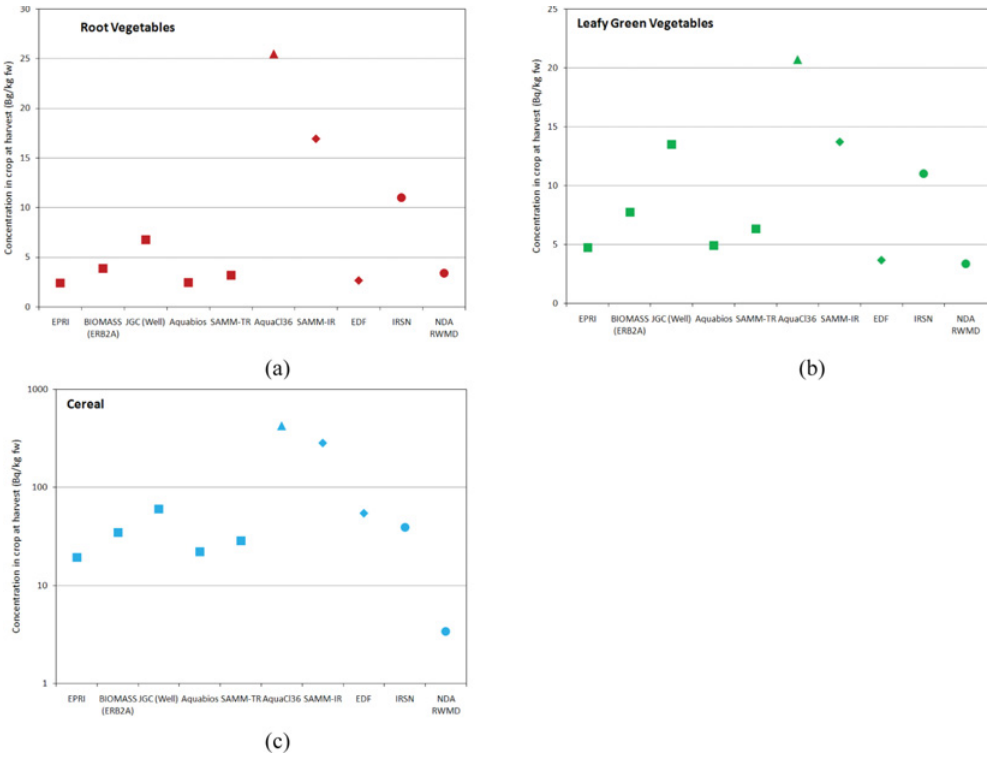

Figure 3. Deterministic calculations of 36 Cl in root vegetables (a), green vegetables (b), and in cereals (c) expressed in $\mathrm{Bq.kg_{fw}^{-1}}$.

The irrigation scenario was as follows:

- There is no irrigation for grass.
- Irrigation will occur only when ET flux > Rain flux.
- Irrigation will occur only when plants are growing in the considered soil (especially for crops).

Contamination from aquifer will occur only when Irrigation flux + Rain flux < ET flux. In this model, the contamination will be upward water movement $*$ 36Cl concentration in groundwater or only a fraction of it.

Considered crop is maize and its Stable Cl concentration is 12.7 10^{-3} $\mathrm{mol.kg^{-1}}$ (Fresh Weight, according to [9] or 9.3 10^{-3} $\mathrm{mol.kg^{-1}}$ (Fresh Weight, according to [13]).

Considered vegetable is lettuce and its Stable Cl concentration is 14.1 10^{-3} $\mathrm{mol.kg^{-1}}$ (Fresh Weight). Considered vegetable is potato and its Stable Cl concentration is 28.2 10^{-3} $\mathrm{mol.kg^{-1}}$ (Fresh Weight).

Grass Stable Cl concentration is 16.9 10^{-3} $\mathrm{mol.kg^{-1}}$ (Fresh Weight).

Considered soil is Khasparov's Greyzem soil. Mean stable chloride concentration in soil is considered to be around 5.6 10^{-3} $\mathrm{mol.kg^{-1}}$ (Dry Weight)

Finally, the organic chloride pool was not considered in this model at its current stage of development. The results presented below assume that 50% of the water taken in as evapotranspiration goes as transpiration, with the other 50% being evaporated into the atmosphere.

4.2 Model Results

We present hereunder the results of this simulation for three kinds of plants

Table 1. Processes considered by the models in the BIOPROTA scenario.

Model Type	Conventional					Specific Activity	Compartmental with isotope ratio approch for plant uptake		More Complex	
Model Name	EPRI	BIOMASS (ERB2A)	JGC (Well)	Aquabios	SAMM-TR	AquaCI36	IR	SAMM-EDF	*IRSN*	*NAD RWMD*
Rainfall				x	x	x	x	x	x	x
Evaporation								x	x	x
Transpiration								x	x	x
Evapotranspiration				x	x		x	x	x	
Sorption of CI-36 to soil	x	x	x	x	x		x	x	x	x
Percolation/leaching/ infiltration to underlying soil layers	x	x	x	x	x		x	x	x	x
Erosion		x	x							
Fartiliser						x	x	x		
Cropping	x	x	x	x					x	x
Irrigation	x	x	x	x	x	x	x	x	x	x

4.3 Model Intercomparison Results

Finally, we present results from an intercomparison study performed under the BIOPROTA international cooperation framework.

The different models considered in this intercomparison exercise were considering different types of processes (see Table 1) and could be incorporated into three types of models:

- Conventional models (BIOMASS (ERB2B), JGL (Soil), SAMM-TR);
- Compartmental models with isotope ratio approach for plant uptake (SAMM-IR, EDF);
- Complex models (IRSN, NDA-RWMD).

Results obtained were quite in accordance: with a factor of 10 for root vegetables and a factor of 6 for green vegetables and if we exclude AquaCL36 and NDA-RWMD models, with a factor of 15 for cereals.

5. CONCLUSION

Chlorine 36 has been identified as one of the radionuclides dominating estimates of annual individual doses to potential exposure groups in post-closure performance assessments of both high and intermediate level radioactive waste depositories. Therefore, modelling its behaviour in the environment is of prime importance. Among all the potential models, the model proposed by IRSN is a dynamic and phenomenological model with a great potential for improvement (for instance taking into account the role of organochlorines). When compared with other models in an intercomparison exercise, it behaves quite well. Moreover, it fits correctly in a bigger structure: the SYMBIOSE programme (SYstemic approach for Modelling the fate of chemicals in the BIOSphere and the Ecological systems) whose main goal is to develop a modelling platform able to sustain an environmental chemical risk assessment.

References

[1] Avila R., R. Thiry Y., Gilbin R., Agüero A., Thorne M., Sheppard M., Tamponnet C., Ikonen A. and Xu S. Recommendations for improving predictions of the long-term environmental behaviour of ^{14}C, ^{36}Cl, ^{99}Tc, ^{237}Np and ^{238}U. (IUR Report 6, IUR Publications, 2006) pp.1–87

[2] Stone J.O., Allan G.L., Fifield L.K. and Cresswell R.G. *Geochim. Cosmochim. Acta* **60** (1996) 679–692.

[3] Huggle D., Blinov A., Stan-Sion C., Korschinek G., Scheffel C., Massonet S., Zerle L., Beer J., Parrat Y., Gaeggeler H., Hajdas W. and Nolte E. *Plant. Space Sci* **44** (1996) 147–151.
[4] Sheppard S.C., Johnson L.H., Goodwin B.W., Tait J.C., Wuschke D.M. and Davison C.C. *Waste Management* **16** (1996) 607–614.
[5] Milton J.C.D., Milton G.M., Andrews H.R., Chant L.A., Cornett R.J.J., Davies W.G., Greiner B.F., Imahori Y., Koslowsky V.T., Kotzer T., Kramzer S.J. and McKay J.W. *Nucl. Instr. and Meth. in Phys. Res*. **123** (1997) 382–386.
[6] Lehmann B.E. and Purtschert R. *Appl. Geochem.***12** (1997) 727–738.
[7] Davis S.N., Moysey S., Cecil L.D. and Zreda M. *Hydrogeol J.* **11** (2003) 217–227.
[8] Corcho Alvarado J.A., Purtschert R., Hinsby K., Troldborg L., Hofer M., Kipfer R., Aeschbach-Hertig W. and Arno-Synal H. *Appl. Geochem*. **20** (2005) 599–609.
[9] Gonze M.A., Garcia Sanchez L., Boyer P., Mourlon C. and Tamponnet C., SPIER3 (International Atomic energy Agency, Vienna, Austria, 2003) pp. 266–277.
[10] Tamponnet C. Modélisation des transferts environnementaux du Chlore 36 (IRSN, Fontenay-aux-Roses, France, IRSN/DEI/SECRE/2006-19,2006) pp. 1–48.
[11] Tamponnet C. Fiches de paramètres pour la modélisation des transferts du Chlore 36 (IRSN, Fontenay-aux-Roses, France, IRSN/DEI/SECRE/2007-20,2007) pp. 1–37.
[12] Limer, L., Albrecht, A., Marang, L., Miquel, S., Tamponnet, C., Nakai, K., Gierzewski, P., Thorne, M. and Smith, G., 2008. Investigation of Cl-36 Behaviour in Soils and Uptake into Crops. (Andra-BIOPROTA N_C.CC.ASTR.08.0048, Chatenay-Malabry, 2008), pp.1–108.
[13] Kashparov V., Colle C., Levchuk S;, Yoschenko V. and Svydynuk N. *J environ Radioact* **94** (2007) 1–15.