Annexe 15

On the use of plants to decontaminate soils around Fukushima-Dai-ichi

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Phytoremediation has been considered to be one of the potential large techniques for the treatment or rehabilitation of hundred of km^2 of $^{134\text{-}134\text{-}137}$Cs-contaminated soils in Japan after the nuclear accident at the Fukushima-Dai-ichi plants. This approach requires various competences, from biotechnology to nuclear safety and bio-engineering, within a global process in order to make phytoremediation both efficient and economically viable.

A - Using plants for decontamination of soils

Phytoremediation can be defined as the use of crops to modulate the content of pollutants in soils. It is a general concept which exploits many useful properties of plants, from "passive" properties (vegetal cover, stabilization, sequestration, regeneration) to "active" mechanisms (rhizofiltration, rhizodegradation, phytoextraction, phytoaccumulation, phytodegradation…), that all help in reducing the impact of pollution.

This approach exploits the basic knowledge on bioaccumulation of radionuclides, in particular of some metal cations, in plant roots and shoots. Advanced methods of molecular biology can be used to improve the selectivity of uptake and monitor the impact on plant physiology. However, former experience with heavy metal polluted soils showed that the knowledge gained in the laboratory cannot be directly extrapolated to the field. The use of European plants species would not be recommended to be used in the Japanese context without a sufficient long testing on site. It would be better to select similar Japanese species in order to select most adapted varieties from local species or crops.

The experience on the field from Chernobyl has confirmed that phytoremediation works, but it also taught that no generic solution exists. Then, specific solutions for the area around Fukushima-Dai-ichi have to be designed. They will take into account the local radioelement profile, the geochemical and geographic features. If a project has to be designed It will take into account the post-harvest processing of contaminated biomass. Conversion of biomass into energy, through heat or biofuels, could be a way to reduce waste volume and to generate some economical feedback.

B -Advantages and limits of phytoremediation

In a general perspective, removal of radioisotopes from soils can be achieved via two main approaches: (i) physico-chemical treatment *in situ* or after excavation, (ii) *in situ* treatment with plants. Promising results have already been obtained using the first approach but the impact on the land is very high since it destroys the pedologic structure of soil. It is restricted to local pollution and may not fit for the majority of agricultural area. In fact, there is no competition between physico-chemical treatment and phytoremediation, the two techniques complement well each other (Table 1). The choice of the most appropriate one will be driven by various criteria, such as soil type, activities and soil usages, environmental impact, time or cost.

	Physico-chemical	Biological
Extent	small areas	large areas
Type of area	highly contaminated	moderately contaminated
Requested time	weeks, months	years
Cost	high	low *
Impact on soils and landscape	local but high (destructive)	null to negligible
Social acceptance	if no other means	green technology

**provided an accessible market for the harvested crops*

***Table 1**. Comparison of the two possible approaches to treat polluted soils. The strengths and weaknesses are analyzed for various important criteria that impact on the process efficiency and sustainability.*

Mechanical removal of radiocontamination may be excluded in some areas for many reasons : economical (low cost-benefit ratio), technical (large volumes or surfaces to treat, incompatibility with ongoing activities) or ecological (increasing land erosion, hydrological impact, vulnerability of the ecosystems…).

In the case of $^{134\text{-}134\text{-}137}Cs$ contaminated soil, the challenge is dual ; i) stabilization of the pollutant in the soil and thus prevention of its migration to aquifers, by growing plants that take only small amounts of radioactivity, ii) removal of pollution, with high loading-radioresistant species.

A large number of studies following the Chernobyl accident have confirmed the efficacy of phytoremediation *(e.g. Rigol et al, 1999, Environ Sci Technol,33,887-895, Entry and Watrud,1998, Water,Air and Soil Pollution,104,339-352).* Incidently, $^{134\text{-}137}Cs$ has also been the matter of several focused reports that evaluated the potential of various types of plants or crops *(Duschenkov et al. 1999 Envir Sci Technol 33, 469-475 ; Putyatin et al, 2006, Radiat Environ Biophys 44,289-298 ; Stauntun et al, 2003, Plants and Soil,254,443-455).* They all confirmed phytoremediation as an alternative method for rehabilitation of large areas *(Duschenkov 2003, Plant and Soil,249,167-175)*, but also show case to case responses; each operation seems unique, indicating that only *in situ* experiments give sense to the efficiency parameters of the process.

When the process is used to transfer a maximum of radionuclides from soil to plant, the final step of the process is frequently eluded. However the harvested biomass must be considered as a radioactive waste, which heavily impacts on the technical complexity and cost of the process

C - A technical proposal

Phytoremediation is suitable for the decontamination of large areas with diffuse radiocontamination if it will be embedded with a large engineering package since an effective phytoremediation program cannot just limit itself to plant cultivation and supply of selected seeds, Its effectiveness also requires both a good knowledge of the site (mapping of the contamination, agronomic management…) and trained local actors (agricultural skills and industrial communities available, compatibility with the national regulations).

Four basic competences have been identified around a global phytoremediation scheme (see boxes in Fig. 1).

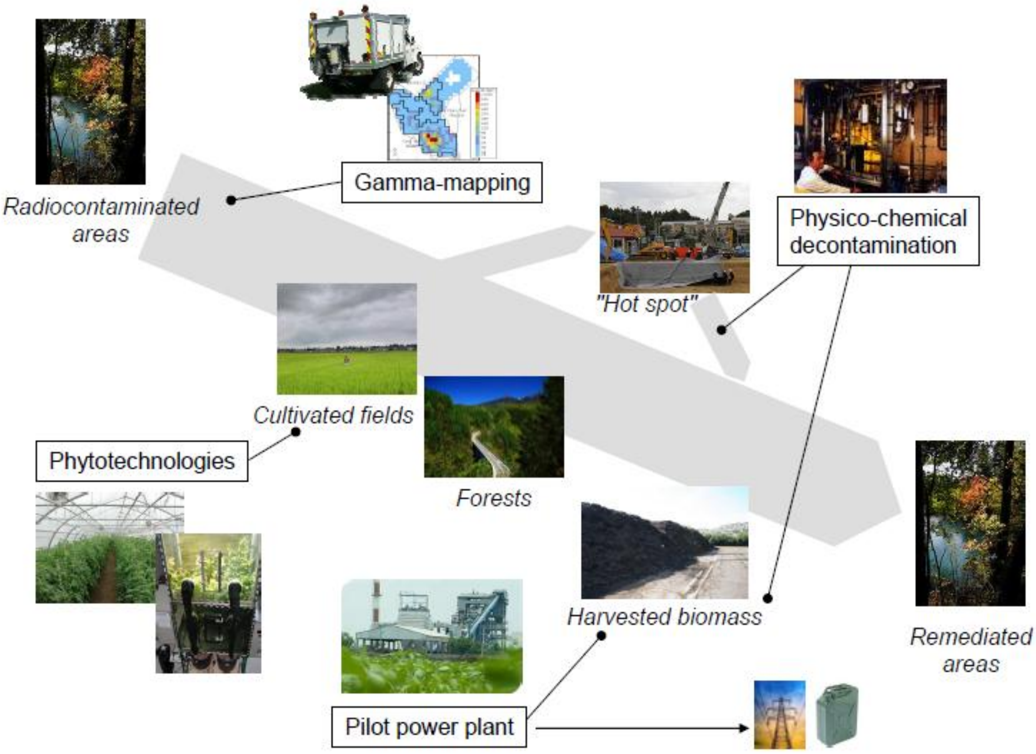

***Figure 1**. Schematic organization of the integrated phytoremediation process. Moderately to low contaminated cultivated fields and fallow lands are primary targets for a phytoremediation program. Forests and local highly contaminated spots can also be addressed if accompanied by physicochemical technologies for decontamination and appropriate engineering.*

Mapping the contaminated areas, nondestructive measurements

Reliable tools for mapping the distribution of gamma-emitting isotopes have been developed, from laboratory vehicles (Fig. 2) to helicopter-borne cameras, and data processing softwares *(De Moura P, Dubot D, Attiogbe J (2011) Radiological evaluation of contaminated sites and soils, VEgAS: an expertise and investigating vehicle. In WM Symposia, Phoenix, USA, 26/2-4/3/2011. Under press*).

The computing developments have resulted to the "Kartotrak" software, in 2003, which is dedicated to the radiological assessment of nuclear sites and contaminated lands *(Dubot D, De Moura P, Attiogbe J (2009) Methodology and tools dedicated to the radiological characterization of contaminated sites, Kick-off meeting of Technical Meeting on the IAEA Network on Environmental Management and Remediation, ENVIRONET, 23-27/11/2009 http://www.iaea.org/OurWork/ST/NE/NEFW/documents/ENVIRONET/kickoff_meeting_2009/ day2/FranceI.pdf).*

***Figure 2.** The VEgAS mobile laboratory. It integrates several gamma spectrometers : a NaI spectrometer (crystal volume 2.4 L, efficiency 30%), and two DSP10 ($^{134-137}$Cs detection limits are 70, 100 and 200 Bq mg.kg^{-1}, depending on vehicle speed, 2.6 km, 5 km and 10 km/h, respectively).*

It would be illusory to try to characterize exhaustively the contaminated area around F-Dai-ichi on all its surface and depth. These mapping resources can be used as follows:

- to characterize the contamination before, during and after remediation;
- to define the optimal size for mapping meshes or samples; help planning the survey strategy;
- to help calculating the costs of survey and remediation.

Physico-chemical decontamination techniques

In addition to solutions for the processing of radioactive liquid wastes, physic chemists have developed several surface decontamination processes based on various products or formulations: gels, foams or powders *(Faure S, Fuentes P, Lallot Y (2008) Vacuumable gel for decontaminating surfaces and use thereof. European patent EP-1941515; Faure S, Fournel B, Fuentes P (2003) Composition, foam and method for surface decontamination. European patent EP-1520279 A2, US patent 20060211592).*

These products, of which the efficiency and specificity are improving over the years, overall aim at reducing the volumes and radioactivity of liquid effluents, or to convert them into solid wastes. Gels and foams may be particularly appropriate for decontamination of small surfaces, for example the roofs and walls of greenhouses or small roads. They can also be used for agricultural machinery.

Another decontamination technology is the use of supercritical CO_2 phase *(Barth F, Bassan S, Lumia G, Perre G (2002) Method, device and installation for cleaning contaminated parts, with a dense pressurized fluid. Worldwide patent WO 0232593, EP 1347840).* It could be used as a solvent to extract chelated radionuclides, e.g. calixarene-chelated $^{134\text{-}137}$Cs. This technology could be advantageously deployed to clean up highly contaminated soil surfaces on-site, such as local "hot spots" (Fig. 3).

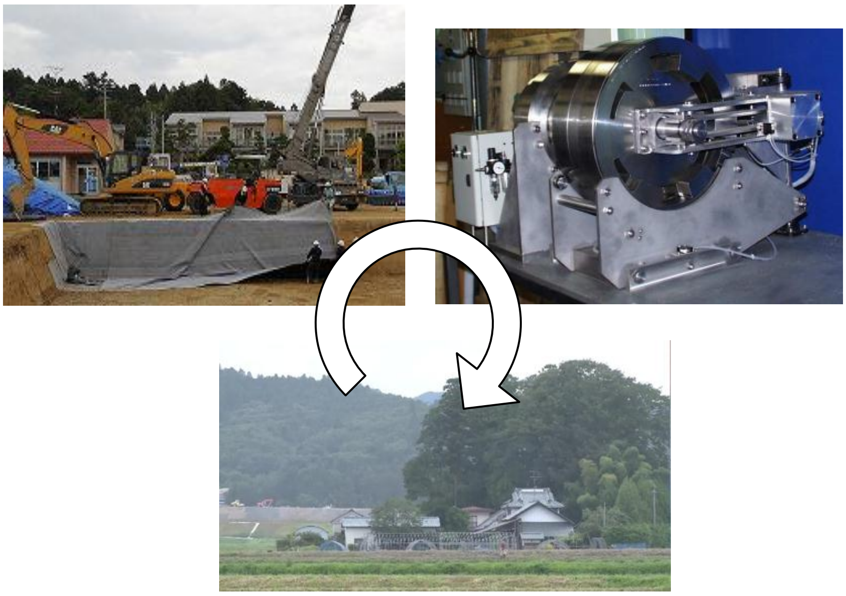

Figure 3. *Treatment of radioactive spots by supercritical CO_2. Contaminated soil is exca-vated, impregnated with a specific chelator for $^{134\text{-}137}$Cs. The complexed radionuclide can then be removed in a pressurized reactor and the treated soil can be put at its original location.*

Supercritical CO_2 is a smooth method in the sense it will not destroy the structure of the material. Regarding biological samples and soils, it is clear that living content will be killed but biochemical features might be preserved, thus facilitating development of microflora.

On the other hand, supercritical water might also be used. It is a strong and destructive oxidizer. It may be interesting for decontamination/degradation of crop residues, including green waste or dead leaves.

Agriculture and forest: crop species, harvesting, storage

Following the Chernobyl accident, many articles described the transfer of radionuclides from the soil to plants and have shown the high variability of transfer coefficients depending on plant species *(Cook et al, 2009, Plant Soil, 324, 169-184)* and genus *(Willey 2010, Radiat Environ Biophys, 49, 613-623)*. An inverse correlation between a high rate of transfer and the biomass produced per surface unit was frequently observed *(White 2003, Plant and Soil, 249, 177-186)*. High yield varieties may content only little quantities of radionuclides in edible parts *(Schneider et al, 2008, Radiat Environ Biophys, 47, 241-252)*.

Reliable data on transfer coefficient values are needed to determine the number of cropping cycles necessary to significantly lower levels of radionuclides in soils. Studies on the phytoremediation of soils contaminated by heavy metals showed no link between the results obtained in the laboratory, or in greenhouse, and those measured in the fields *[Vavasseur A. (2009) Bioremediation in Toxicologie nucléaire environnementale et humaine (Menager MT, Garnier-Laplace J, Goyffon M, eds). Lavoisier, Paris. ISBN 2-7430-1174-2 (in French)]*. Then, access to actual plots of land is indispensable to select appropriate crops.

The performance of plants can largely be explained from biochemical or physiological properties and can be dissected or modulated by genetic engineering to study molecular transporter of metals and their regulations *(Song WY, Sohn EJ, Martinoia E, Lee YJ, Yang YY, Jasinski M, Forestier C, Hwang I, Lee Y (2003) Engineering tolerance and accumulation of lead and cadmium in transgenic plants. Nature Biotechnol., 21, 914-919. - Migeon A, Blaudez D, Wilkins O, Montanini B, Campbell MM, Richaud P, Thomine S, Chalot M (2010) Genome-wide analysis of plant metal transporters with an emphasis on poplar. Cell. Mol. Life Sci., 67, 3763-3784.)*, or the cellular mechanisms of the radioelements storage *(Isaure MP, Fraysse A, deves G, Le Ray P, Fayard B, Susini J, Bourguignon J, Ortega R (2006) Micro-chemical imaging of cesium distribution in Arabidopsis thalian plant and its interaction with potassium and essential trace elements. Biochimie, 88, 1583-1590)*. For example, plants expressing a high level of transporter activity might display a higher potential for accumulating radionuclides (Fig. 4).

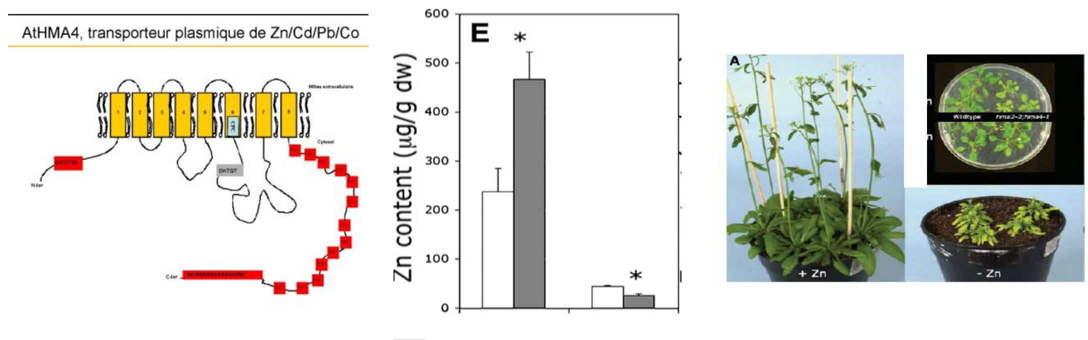

Figure 4. *Regulation of AtHMA4 to control metal uptake by plants. Left panel, schematic molecular structure of the AtHMA4 protein. It is important in the transmembrane flux of a large spectrum of divalent metal cations Zn^{2+}, Cd^{2+}, Pb^{2+} and Co^{2+}. Right panels, example of functional consequences: overexpression of this protein in plant roots increased the overall Zn content, while inactivation of the gene gave rise to a dramatically lowered metal content.*

Regarding the important role of speciation for plant transfer, the recent study performed by a Japanese team[#] that analyzed the *"relationships among ^{137}Cs, ^{133}Cs, and K in plant uptake observed in Japanese agricultural fields"* (Kamei-Ishikawa et al. 2011. *J Radioanal Nucl Chem,* available on line), is a very valuable information which was interestingly submitted on January 2011, two months before the Great Tohoku Earthquake.

In areas covered with perennial species (orchards, forests) that received fallouts, lowering the radioactivity levels requires the tedious harvesting of branches, leaves, trunks that would have otherwise contaminate soils. Forestry machineries could be engaged to harvest and transport this ligno-cellulosic biomass in compliance with radioprotection guidelines. Finally, as well in agricultural as in forest production, very large amounts of biomass will be harvested and their storage gives rise to nuclear safety issues.

From radioactive wastes to energy supply

Year after year, the harvested radio-contaminated biomass will represent considerable volumes. This biomass can be used as a renewable energy source. Several technologies are envisioned:

- combustion in a high pressure boiler; this large scale technology based on water vapor to directly produce electricity would be able to process a minimum of 50,000 tons of biomass per year;
- gasification of biomass. This technology is still under development but is considered as a promising route to produce CO_2-neutral fuels, either liquid (Diesel Fischer-Tropsch, methanol…) or gaseous (Synthetic Natural Gas, "Bio-SNG"). Large scale industrialization of this process is planned on a short to mid-term time scale, depending on the process options. The building of demonstration plants based on the Biomass-to-Liquid process (BtL) has already begun in Germany and France *(Kiener C, 2008, in 16th European Biomass Conference, Valencia - Dupont C, da Silva Perez D, Labouchée C, Rougé S, Graffin A, Mithouard JC, Berthelot A, Labalette F, Pitocchi S, (2010) Bioenergy II: suitability of wood chips and various biomass types for use in plant of BtL production by gasification. Intern J Chem React Engin, 8, A74, 21p).*

Both technologies are suitable with the requirements of radioprotection guidelines. The French experience of SOCODEI (http://www.socodei.fr/en/waste-processing/) with selective filters, to catch radionuclides during combustion and prevent their dispersion in the environment, might be very helpful. The residual ashes will have to be managed as small volumes/high activity wastes.

This combination of phytoremediation with bio-energy supply is a direct way (i) to eliminate large volumes of contaminated biomass, (ii) to create a new industrial activity in the sinister area and (iii) to supply energy for decontamination/reconstruction duties. A preliminary methodological paper, issued from the European project RECOVER, gives a first area on the economic viability of this production *(Vandenhove H, Goor F, O'Brien S, Grebenkov A, Timofeyev S, 2002, Biomass and Bioenergy,22,421-431)*. This study is 10 years old and deserves to be actualized for the current context in Japan.

[#] Nao Kamei-Ishikawa (Iwate University), Keiko Tagami and Shigeo Uchida (NIRS Chiba).