Remediation of Areas Contaminated by Caesium: Basic Mechanisms Behind Remedial Options and Experience in Application

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Abstract There are many areas around the world contaminated with radiocaesium to different extents due to global fallout from nuclear weapons testing, radiation accidents or inadequate waste disposal practices. In recent decades, a wide range of options for remediation of these areas have been developed, tested and implemented to mitigate the potential doses in such areas. A large amount of data on the effectiveness of remediation options has been generated, together with information on ancillary factors such as technical feasibility and side effects. The chapter aims to provide information on available options for remediation of terrestrial and freshwater ecosystems contaminated with radiocaesium. An associated objective is to provide scientific information on the basic mechanisms which impact on effectiveness of the described remedial options.

Keywords Radiocaesium • Terrestrial and freshwater environments • Remediation • Remedial options

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1 Introduction

Remediation (or remedial action) is normally defined as any measure that intended to reduce radiation exposure from existing contamination through actions applied to the contamination itself (the source) or to the exposure pathways to humans (IAEA 2014). A remediation strategy can be defined as a time-dependent sequence of remedial actions which have been justified and are undertaken in a specified area for a defined time. Although the main objective of a remediation action within a remediation strategy is to reduce or to prevent radiation doses to humans, the provision of reassurance to the population of the contaminated areas is also an important objective of remediation as it helps to maintains public confidence in areas affected by radioactive contamination (Fesenko and Howard 2012).

During ^{235}U fission, 13 isotopes of Cs are formed. Among them, the radioactive isotope ^{137}Cs is of greatest importance, as it has a large fission yield (6.2 %) and a relatively long physical half-life (30.17 years). Caesium isotopes are characterised by high biological mobility, because caesium is an alkali element and a chemical analogue of a biochemically important element K (Nisbet and Woodman 2000 2000; Simon et al. 2002). The dominant aqueous species in soil and aquatic systems is thought to be free Cs^{+}. An important feature of the behaviour of Cs isotopes is their ability for non-exchangeable sorption (fixation) by the soil solid phase (Smolders and Merckx 1993). The Cs^{+} ion forms only extremely weak aqueous complexes with SO_4^{-2}, Cl^{-}, and NO_3^{-}, and thus the formation of inorganic complexes is not believed to be a major influence on caesium speciation (Yudintseva and Gulyakin 1968). Cs becomes associated with the clay mineral fraction of soils (Sawney 1964) with a high selectivity. Accumulation of ^{137}Cs in plants depends on soil properties with the concentrations of competing cations exerting a modifying effect. Cs mobility is relatively high in peat and sandy soils (Sanzharova et al. 1996). Remediation of areas affected by radiocaesium is focused on reduction of both ingestion dose from the consumption of contaminated foodstuffs or drinking water and external dose from surfaces contaminated by deposited radionuclides. Inhalation of resuspended material is normally a negligible pathway and its importance is much lower than that from ingestion and external exposure in most contamination scenarios.

A large number of remedial actions to be applied in areas affected by radiocaesium have been developed and applied, especially when there has been significant contamination arising from releases such as the Chernobyl accident (Alexakhin 2009) in 1986, the incident in Goiânia, in 1987 in Brazil (Eisenbud, and Gesell 1997) and recently, the Fukushima Daiichi accident (IAEA 2015). Therefore, in recent decades many remedial options have been developed, tested and implemented in the contaminated areas. As a result, a large amount of data on the effectiveness of various remediation options has been generated, together with information on ancillary factors such as the required resources and costs. The experience gained has been invaluable in quantifying the efficiency of remedial actions. In addition, prominence has been given to identifying many other factors which affect the potential application of various remediation options such as environmental conditions, radionuclide properties, land use of the affected areas and response from the local population and stakeholders which can affect the effectiveness and impact on the suitability of remediation options (Howard 2000; Fesenko et al. 2000a, b, 2012). Information on effectiveness is often presented as a reduction factor i.e. reduction of radiocaesium concentration in the product or reduction of internal and external dose after application of the remedial option. Alternatively, a percentage reduction in radionuclide concentration in the target medium (i.e. soils and crops) after implementation is reported. The latter is most often used for food treatment options and has, therefore, been used here for these options.

2 Basic Mechanisms Behind Soil-Based Remedial Options Affecting Radiocaesium Mobility

Soils constitute the main long-term reservoir of radionuclides in terrestrial ecosystems. Many remediation options are applied to the soil aiming to modify the soil parameters that affect radionuclide mobility. Soil-based remediation options can be divided into those that alter the soil structure (mechanical treatments) and those that directly modify the chemical characteristics of the soil. The application both of these measures has the advantage that they are easy to implement in most areas as the necessary equipment and expertise is already available. The mechanisms behind soil-based remedial options affecting radiocaesium mobility discussed in this section are illustrated based on data obtained for six case studies: two from Belarus (Dublin and Sawichi), two from Russia (VIUA and Rudnuy) and two from Ukraine (Matevki and Christinovka) located in the areas affected by the Chernobyl accident. A full description of these sites is given elsewhere (Vidal et al. 2000).

2.1 Factors Governing Sorption on Cs in Soils

Cs soil sorption properties vary considerably and are commonly quantified using both the Radiocesium Interception Potential (RIP) and the K and NH_4^+ concentrations in the exchange complex of the soil solid phase and soil solution (Sweeck et al. 1990; Vandebroek et al. 2012). The RIP estimates the capacity of a given soil to specifically sorb Cs and can be readily determined based on routine laboratory experiments (Wauters et al. 1996a). The most common protocol to determine the RIP is based on pre-equilibrating the samples with a solution containing 100 mmol L^{-1} of Ca and 0.5 mmol L^{-1} of K (m_K). After pre-equilibrating the samples, these are equilibrated with a solution with the same K and Ca composition, but labelled with radiocaesium. The distribution coefficients (K_d (Cs)) are obtained by measuring the radiocaesium activity concentrations in the supernatant, before and after the equilibration. The calculated product K_d (Cs) × m_K defines the RIP value (in mmol kg^{-1})

(Wauters et al. 1996a). The RIP value is related to the content and selectivity of expandable clays, especially illite, vermiculite and other 2:1 phyllosilicates, in which Frayed Edge Sites (FES), which are specific sites for Cs sorption, are present (Sweeck et al. 1990). Other exchange sites are of little relevance for Cs sorption (Brouwer et al. 1983; Cremers et al. 1988; Vidal et al. 1995), except for soils with an extremely low clay content (such as soils with a organic matter content over 90 % or highly sandy podzols). In such soils, the role of regular exchange sites (RES) should be taken into account (Vidal et al. 1995; Rigol et al. 1998).

As Cs sorption is controlled by the FES, the Cs solid-liquid distribution coefficient at these sites (K_d^{FES} (Cs)) accounts for more than 80 % of the total sorption process (Vidal et al. 1995). The K_d^{FES} (Cs) can be predicted by dividing the RIP value by the sum of K and NH_4^+ concentrations in the soil solution, the latter amplified by the NH_4-to-K trace selectivity coefficient in the FES (K_c^{FES} (NH_4/K)) (Sweeck et al. 1990). This parameter, which can be easily quantified by laboratory experiments, ranges from 4 to 8 for soils in which specific sites control Cs sorption quantitatively, and down to 2 in those soils where sorption occurs at regular exchange sites (Wauters et al. 1996b; Rigol et al. 1998). For a more accurate prediction of the value of K_d (Cs), a second term must be added to account for Cs sorption at regular exchange sites (RES) (K_d^{RES} (Cs)), by dividing the sum of the exchangeable K and NH_4^+ by the sum of K and NH_4^+ concentrations in the soil solution (in mmol L^{-1}), assuming a selectivity coefficient NH_4/K of approximately 1 at these sites. The overall equation incorporating sorption at specific and regular sites may be written as follows:

$$K_d(Cs) = K_d^{FES}(Cs) + K_d^{RES}(Cs) = \frac{RIP}{K_{ss} + K_c^{FES}(NH_4/K) \bullet NH_{4ss}^+} + \frac{K_{exch} + NH_{4exch}^+}{K_{ss} + NH_{4ss}^+}$$

For the case of highly saline soils, near to marshlands, with high *Na* concentrations in the soil solution, the equation may be slightly modified to include the potential competitive role of Na and its effect on the quantification of K_d^{FES}, correcting the Na concentration by the Na-to-K trace selectivity coefficient in the FES, K_c^{FES} (Na/K). As this coefficient has values of around 0.02 (Wauters et al. 1996b), the role of Na will only have a significant effect when there is an unusually high Na concentrations. The equation may be simplified by considering that Na and NH_4^+ concentrations are generally much lower than K concentrations, as is the case for most agricultural systems with mineral soils. As the value of K_d^{FES} (Cs) is much larger than the value of K_d^{RES} (Cs), K_d (Cs) is reasonably well predicted by the following equation, except for those soil types (upland, peat soils; soils affected by flooding) in which $NH_4^+{}_{ss}$ can be significant:

$$K_d(Cs) = \frac{RIP}{K_{ss}}$$

To date, attempts to predict the RIP value based on soil properties have only been partially successful. Waegeneers et al. (1999) showed that the RIP value depended not only on the clay content, but also on the type of clay and geological origin of the soil. In their study involving a stepwise regression analysis to two sets of soils, the clay content alone accounted for up to 71 % of the variance of the RIP in the most favourable set of soils, whereas for another set of soils it explained only 13 % of the variance. The regression improved when the silt content and the pH were added to the clay content, depending on the origin of the soils. In more recent work, Gil-García et al. (2009) showed that clay content alone explained 40 % of the variance of the RIP values, and that there was a good correlation between the two variants [$log\ RIP = 1.8 + 1.0 \times log\ clay$; $r = 0.64$; $n = 108$], while the inclusion of the silt content explained the 60 % of the total variance [$log\ RIP = 1.5 + 0.9 \times log\ clay + 0.45 \times log\ silt$]. When organic soils were excluded from these analyses, the clay content alone explained up to 57 % of the variance, with an improved correlation [$log\ RIP = 2.1 + 0.95 \times log\ clay$; $r = 0.76$; $n = 86$], while the inclusion of the silt content increased the explained variance to up to 62 % [$log\ RIP = 1.6 + 0.8 \times log\ clay + 0.5 \times log\ silt$] (Gil-García et al. 2009). Therefore, although the RIP values can be partially predicted from clay and silt contents, limitations to properly predict RIP from soil properties suggests the need to quantify the RIP for a better estimation of the K_d (Cs) (Gil-García et al. 2011). In agreement with the K_d(Cs) dependence, the CR_{ss}(Cs) (soil solution–plant concentration ratio[1]) is expected to vary inversely with K_{ss} and NH_{4ss}. However, for $K_{ss} + NH_{4ss}$ values higher than 0.5–1 mM, the CR_{ss}(Cs) remains reasonably constant (Smolders et al. 1997a, b; Camps et al. 2004). Therefore, a major increase in NH_{4ss} may lead to an increase in F_v(Cs) due to the different mechanisms affecting changes in the K_d (Cs) (the NH_{4ss} is multiplied by the $K_C^{FES}(NH_4/K)$) and in the CR_{ss}(Cs) (in which NH_{4ss} has the same weight as K_{ss}) (Vidal et al. 1995).

2.2 *Mechanisms Related to Mechanical Treatments*

Radionuclide migration in soil is a relatively slow process in most types of soil and radionuclides deposited on the soil surface typically remain in the upper soil horizons for many decades. Mechanical treatments are intended to decrease the pool of radionuclides in the rooting zone by dilution achieved by mixing the contaminated topsoil layer with deeper soil layers which have lower radionuclide contents. Mechanical treatments such as shallow, deep or skim and burial ploughing, will have the following positive effects:

- dilution, leading to a lower radionuclide activity concentration in crops;
- transfer of radionuclides down to a soil horizon below the crop rooting area;
- decrease in the resuspension of contaminated soil;
- decrease in the adhesion of contaminated soil to plants and
- reduction of external dose rate.

[1] Soil solution–plant concentration ratio, CR_{ss}, is the ratio of the radionuclide activity concentration in the plant (Bq kg^{-1} plant (dw)) to that in the soil solution (Bq L^{-1}).

Mechanical treatments may also modify the capacity of soils to immobilize radionuclides. Examples include situations when the radionuclide-contaminated layer is mixed with a soil layer of different mineralogical composition, or when soil loosening causes a change in the amount of sorbing surface which determines the extent of binding of radionuclides. To illustrate this phenomenon, Table 1 provides RIP values for a ploughed layer, in comparison with *RIP* values of undisturbed 0–10 cm and 10–20 cm layers soils of five sites (Camps et al. 2004). At the VIUA and Rudnuy sites, a decrease in the RIP values in the ploughed soils was noted due to the presence of a 10–20 cm leached horizon with poor radiocaesium sorption properties. This was shown by the lower RIP values in the 10–20 cm layer than those of the 0–10 cm depth layer. The decrease in RIP values suggests there would be a detrimental effect on radiocaesium mobility, since the resulting ploughed soils would have a lower specific sorption capacity in the rooting area. At sites in Dublin and Sawichi there were similar *RIP* values in unploughed control and ploughed soils no secondary detrimental effects on the *RIP* due to ploughing were observed. At the Mateyki site an increase in the radiocaesium sorption properties was observed due to a heterogeneous soil profile in which the topsoil had a lower specific sorption capacity than that of deeper horizons. At this site, the 10–20 cm soil layer in the unploughed control plot was highly heterogeneous in clay content, with a wide range of *RIP* values, which were often greater than those in the topsoil. Thus, ploughing created a soil profile, with higher *RIP* values than the original soil before ploughing in the 0–10 cm. The data show that ploughing not only redistributes radiocaesium in soil but also may change the top soil sorption properties affecting caesium mobility in the soil and root uptake by plants.

Table 1 Changes in RIP after ploughing undisturbed soils

Soils and treatments	*RIP* (mmol kg^{-1})	Soils and treatments	*RIP* (mmol kg^{-1})
Sawichi		*Mateyki*	
C 0–10	240	C 0–10	70
C 10–20	185	C 10–20	15–810
Ploughed layer	150	Ploughed layer	140
Dublin		*VIUA*	
C 0–10	87	C 0–10	90–485
C 10–20	75	C 10–20	27
Ploughed layer	80	Ploughed layer	39
Rudnuy			
C 0–10	545		
C 10–20	39		
Pl	190		

2.3 Mechanisms Related to Changing the Chemical Characteristics of Soils

The radionuclide activity concentration in plants depends on that in the soil solution and on the ion uptake process. The concentration of radionuclides in the soil solution depends on the total concentration in the solid phase, the radionuclide solid–liquid distribution coefficient (K_d)[2], and the reversibly sorbed fraction (Vidal et al. 2009). For many radionuclides, after some time has elapsed since sorption, the reversibly sorbed fraction will be of a similar order of magnitude for soils with similar properties. Thus, the range of variation should be narrower than the range of variation of K_d for all soils, and, therefore, radionuclide availability is often quantified solely in terms of the K_d. The ion uptake process from soil solution to the plant includes plant physiological aspects, related to nutrient uptake and selectivity. Therefore, (CR_{ss})[3] is assumed to depend on the activity concentrations of radionuclide competitive species in the soil solution. Thus, the soil-plant concentration ratio (F_v)[4] may be described by the following relationship: $F_v = CR_{ss}/K_d$. Therefore, the F_v values decrease with increases in the K_d and/or with decreases in the CR_{ss} (Vidal et al. 2009). The same affect is observed also for T_{ag} (aggregated transfer factor defined as the mass activity density (Bq kg^{-1}) in the plant per unit area activity density, A_a (Bq m^{-2}) in the soil m^2 kg^{-1}(ICRU 2001).

2.4 Effects of Commonly Applied Agricultural Treatments on K_d and T_{ag}

The application of potassium to soils is most effective in reducing F_v(Cs) when the K_{exch} is less than 0.5 meq 100 g^{-1} soil, that is, for K_{ss} in the micro-molar range (lower than 0.5–1 mM). Over this range, additional doses of K fertiliser may have a negative effect on T_{ag}(Cs) transfer, because the decrease in the K_d(Cs) is not compensated for by an increase in the dilution effect in the soil solution, and then in the CR_{ss}(Cs) (Smolders et al. 1997a, b; Camps et al. 2004). Liming may be an effective option for reduction of Cs transfer to plants because in soils with an optimum K_{ss} (over 1 mM), the F_v(Cs) may decrease due to competition between Cs and Ca + Mg for exchange sites in the apoplast of the root cortex (Smolders et al. 1997a, b). Also, the increase in $(Ca + Mg)_{ss}$ may increase the K_d(Cs) in soils, due to the masking of RES, an

[2] Distribution coefficient is the ratio of the mass activity density (A_m, in Bq kg^{-1}) of the specified solid phase (usually on a dry mass basis) to the volumetric activity density (A_v, in Bq L^{-1}) of the specified liquid phase.

[3] Soil solution–plant concentration ratio, CR_{ss}, is the ratio of the radionuclide activity concentration in the plant (Bq kg^{-1} plant (dw)) to that in the soil solution (Bq L^{-1}),

[4] Concentration ratio, Fv, is the ratio of the radionuclide activity concentration in the plant (Bq kg^{-1} dw) to that in the soil (Bq kg^{-1} dw).

expansion of clay interlayers, or a decrease in the concentration of monovalent species in the soil solution (Rigol et al. 1999a). The effect of the use of agricultural treatments, combined with ploughing, on changes in the soil properties and related changes in Cs transfer is illustrated in Fig. 1 which shows changes in ^{137}Cs transfer to plants in a set of soils submitted to different agricultural practices, such as ploughing (Pl), NPK fertilization (NPK) and liming (L) (Camps et al. 2004).

In the first year of the experiments, ploughing (Pl) plus reseeding was the most effective treatment in decreasing radiocaesium transfer to plants in most sites (see

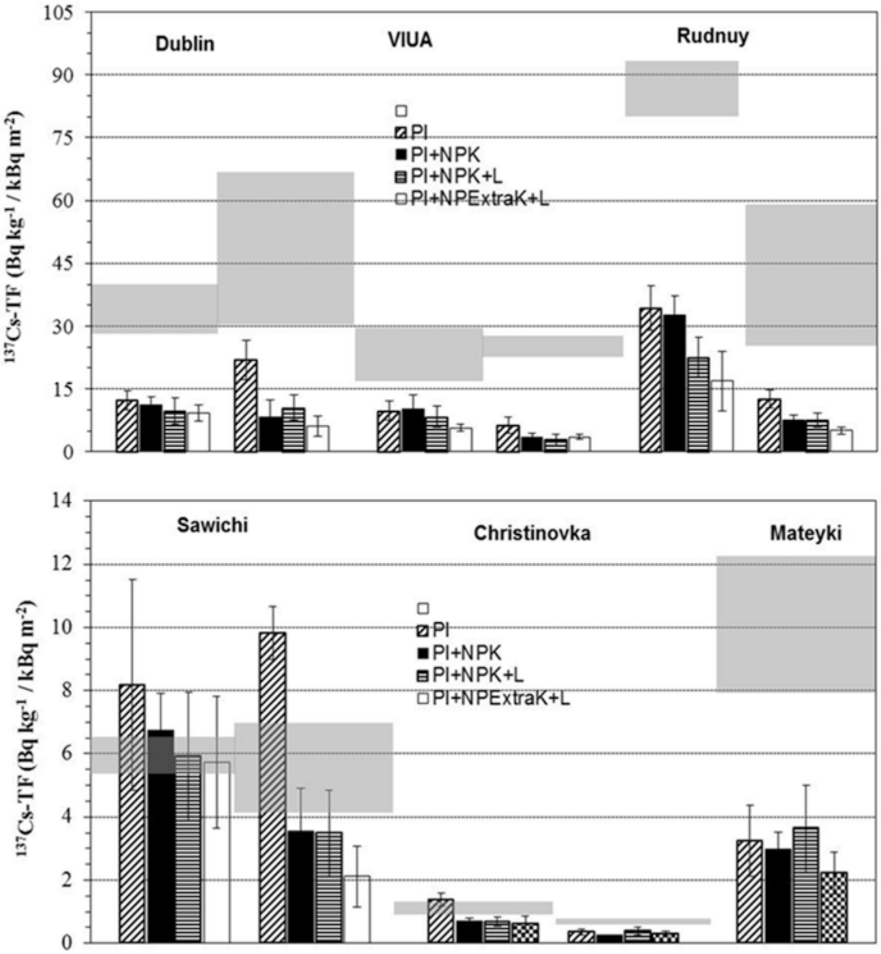

Fig. 1 Effect of agricultural practices on ^{137}Cs aggregated transfer factor (T_{ag}) at the test sites. *Error bars* indicate one standard deviation. Control data with normal application rate of fertilizers are represented by a *hatched area* that includes the mean ± one standard deviation

Mateyki, Dublin, VIUA and Rudnuy sites), while at the Sawichi and Christinovka sites similar or even higher T_{ag} (Cs)[5] values were measured in ploughed (Pl) than in unploughed control plots. The application of NPK fertiliser caused a further decrease in radiocaesium transfer at most sites, especially when applying the extra dose of potassium (Fig. 1), with the exception of the Sawichi site. The effectiveness of liming was minor. Treatment effects became generally more pronounced in the second year. However, the T_{ag}(Cs)s of control soils also decreased with time at some sites (Christinovka and Rudnuy). Key parameters explaining the effect (or lack of) of these treatments on Cs transfer are the potential changes in potassium and NH_4^+ status in the soil exchangeable complex and in the soil solution due to the agricultural treatments. Table 2 summarizes the data related to potassium concentrations in the exchange complex and potassium concentration in the soil solution in the unploughed (control) and the treated plots. As stated above, for potassium concentrations in the soil solution in the micromolar range (lower than 1–2 mM), an increase in the potassium concentration would lead to a decrease in radiocaesium root uptake. Over 1–2 mM, additional application of potassium fertiliser may have a negative effect on radiocaesium transfer, because the decrease in the K_d (Cs) is not compensated for an increase in the dilution effect in the soil solution, and then in the T_{ag}(Cs) (Smolders et al. 1997a, b).

In the first year of the experiments, there was a general increase in the potassium concentrations at several sites, especially in the soil solution, after the addition of extra doses of the NPK fertiliser (see Sawichi, Dublin and VIUA sites), achieving in some cases potassium concentrations in the soil solution in the range of 1–2 mM. At some of the sites, the fertiliser supply was insufficient to increase potassium concentrations over those found in unfertilised ploughed plots, with values remaining in the initial micromolar range (see Christinovka and Mateyki sites). These findings agree with the transfer pattern observed in Fig. 1. The optimum potassium concentrations in soil solution achieved at several sites could not be maintained due to a potassium depletion caused by the increase in nutrient root uptake resulting from the plant growth, or from the increase in biomass in the second year of experiments (compare first and second year at the Dublin and Sawichi sites). The data imply that the application of higher amounts of potassium fertiliser would have maintained an optimum potassium concentration in the soil solution even after a higher plant yield in more mature meadows. The outcome would have been a further decrease in radiocaesium transfer to the crop.

2.5 *Effects of the Application of Organic and Mineral Materials on K_d and T_{ag}*

Application of natural organic and mineral materials to soils is a standard agricultural practice in some regions. The addition of such amendments, usually applied with fertilizers, may improve the agrochemical properties of the soil, leading to an

[5] T_{ag} (Cs) is the ratio of the Cs activity concentration in plant (Bq kg^{-1}) divided by the total deposition on the soil (Bq m^{-2})

Table 2 Effect of agricultural practices on the RIP values, the exchangeable K (K_{exch}, $cmol_c\ kg^{-1}$) and K concentration in the soil solution (K_{ss}, $mmolL^{-1}$)

	$K_{exch,}$ 1st year	2nd year	K_{ss}, 1st year	2nd year
Sawichi				
C 0–10	0.24	0.33	1.7	1.6
C 10–20	0.06	nd	0.3	nd
Pl	0.17	0.07	0.6	nd
Pl+NPK	0.22	0.12	1.0	0.40
Pl+NPK+L	0.30	0.14	1.4	0.47
Pl+NP1.5 K+L[a]	0.30	0.19	1.8	0.60
Dublin				
C 0–10	0.80	0.70	0.8	1.2
C 10–20	0.30	nd	0.4	nd
Pl	0.50	0.50	0.9	nd
Pl+NPK	0.80	0.36	1.4	0.24
Pl+NPK+L	0.68	0.30	1.4	0.35
Pl+NP1.5 K+L	0.82	0.37	1.6	0.43
Christinovka				
C 0–10	0.18	nd	0.10	nd
Pl	0.19	0.18	0.10	0.02
Pl+NPK	0.21	0.24	0.16	0.04
Pl+NPK+L	0.24	0.21	0.13	0.025
Pl+N2P2K+2 L	0.25	0.20	0.20	0.033
Mateyki				
C 0–10	0.08	na	0.06	na
C 10–20	0.02	na	0.013	na
Pl	0.13	na	0.06	na
Pl+NPK	0.16	na	0.10	na
Pl+NPK+L	0.14	na	0.07	na
Pl+N2P2K+2 L	0.11	na	0.08	na
VIUA				
C 0–10	0.12	0.08	1.1	0.4
C 10–20	0.06	nd	nd	nd
Pl	0.07	0.09	0.5	0.4
Pl+NPK	0.09	0.07	0.4	0.8
Pl+NPK+L	0.12	0.07	0.4	0.6
Pl+NP1.5 K+L	0.11	0.10	1.1	0.5
Rudnuy				
C 0–10	0.30	0.20	0.9	0.7
C 10-20	0.05	nd	nd	nd
Pl	0.14	0.13	0.4	0.26
Pl+NPK	0.14	0.14	0.4	0.36
Pl+NPK+L	0.17	0.16	0.4	0.5
Pl+NP1.5 K+L	0.17	0.23	0.5	1.1

Notation of $Pl+{}_xN_XP_yK+L$ means combine application of ploughing mineral fertilizer and lime. x is an increase of application rate in relation to the normal one
nd Not detected, *na* not analysed

increase in crop yields, and possibly also enhancing soil sorption properties (Nisbet 1993; Nisbet et al. 1997). The application of these sorbent materials to the soil modifies the soil solid phase which influences radionuclide uptake in two ways: (1) by increasing the sorbing capacity for radionuclides, and (2) by modifying the composition of the soil solution. To be effective such materials must increase the radionuclide sorption potential (or K_d) of the soils. For Cs, this can readily be achieved by increasing the RIP in the soil. Because only low doses of amendments can be used for an economically justifiable remediation strategy, the K_d of radiocaesium in the added material should exceed the normal values in the soils to be treated by several orders of magnitude to have a significant effect at field level. Therefore, the sorption characteristics of the materials have to be determined in the laboratory before being used at field level (Valcke et al. 1997; Camps et al. 2003). Materials with high mineral matter content, enriched in 2:1 phyllosilicates, are the most suitable materials for sorbing radiocaesium. The addition of organic material would lead to an increase in the (1) organic matter in soil, (2) contents of nutrients and microelements, (3) CEC and (4) $(Ca+Mg)_{exch}$. These increases may lead to a lack of effect of the treatment on radiocaesium sorption or even a detrimental effect in reducing F_v(Cs). A case study, using field plots from Ukraine, Belarus and Russia, is used here to illustrate how the addition of materials does not necessarily lead to a decrease in radiocaesium transfer to plants (Vidal et al. 2001; Camps et al. 2003). Table 3 presents a summary of the main general characteristics of the soils and amendments used in this study.

Figure 2 provides data on the changes of T_{ag}(Cs) for the study sites mentioned earlier, including both the control (hatched area) and amendment plots (vertical

Table 3 Characteristics of soils and amendments tested

Soil amendments	pH	OM	CEC	K_{exch}	NH_{4exch}	Sand	Clay	Texture
Sawichi								
Control	4.4	7.9	25	0.26	0.22	72	5.3	Sandy-loam
Sapropel	4.8	38	86	0.59	0.92	47	27	Sandy-clay loam
Turf	5.9	45	85	0.44	2.1	63	0.7	Sandy-loam
Dublin								
Control	6.3	78	140	0.68	0.50	nd	1.3	–
Mineral soil	6.5	0.1	8.2	0.08	0.08	83	0.6	Loamy-sand
Mateyki								
Control	6.4	44	57	0.24	0.08	nd	12	–
Sandy soil	5.9	0.2	4.4	0.06	0.08	90	1.6	Sandy
Loamy soil	6.1	0.8	10	0.14	0.08	55.9	12.9	Sandy-loam
VIUA								
Control	6.7	3.5	9.7	0.12	0.20	79.8	3.4	Loamy-sand
Rudnuy								
Control	5.6	13	15	0.13	0.11	90.2	3.1	Sandy
Phosphorite	6.8	2.4	8.6	0.40	0.09	26.9	20.1	Loamy-silt
Polygorskite	7.7	4.3	12	0.41	0.08	1.4	81.1	Clay

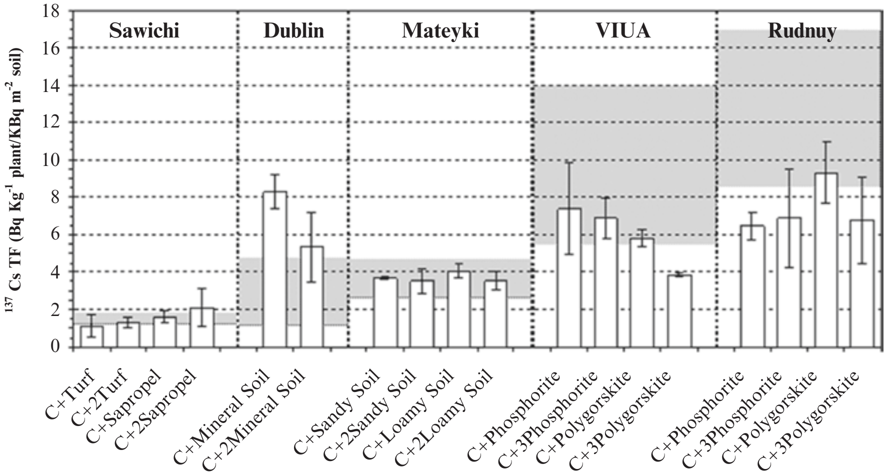

Fig. 2 Effect of soil amendment addition on ^{137}Cs aggregated transfer factor for plants Tag. *Error bars* indicate one standard deviation. Control treatments are represented by a *hatched area* that includes the mean ± one standard deviation

bars) (Vidal et al. 2001). The addition of amendments did not lead to a decrease in transfer in most of the treatments. A positive effect of phosphorite and especially polygorskite amendments was determined at only at a few sites. Radiocaesium transfer to the plants was reduced by around 1.5–2-fold at the VIUA site, where the highest amount of polygorskite was used, and at the Rudnuy site, where two amendments were applied. At the Dublin site, a negative effect was observed when using the mineral amendment, whereas at the Mateyki site a negligible effect was observed for both amendments tested.

RIP values were determined in control and amended soils (Table 4). Although RIP values were highly variable, the RIP values of the amended soils seemed to increase in only a few scenarios. This was the case at the Mateyki site when adding the highest amount of loamy soil, and at the Rudnuy site when adding polygorskite clay and phosphorite. The improvement in the RIP values was related to the much higher values of the RIP of the amendment related to the untreated soils. The low doses prevented a significant, positive effect in increasing the RIP and then reducing radiocaesium transfer in most cases, although the increase in RIP values at the Rudnuy site when phosphorite and polygorskite were applied corresponded to the decrease in radiocaesium transfer observed at field level. The Dublin site constituted a different scenario because the RIP values decreased when adding the mineral material, as the mineral soil added had a sandy texture, with lower clay content than the Dublin soil. This amendment also had a lower affinity for radiocaesium, with a lower RIP than that of the Dublin control soil. The resulting decrease in RIP may explain the increase in radiocaesium transfer to the plants observed at this site. The effect of the amendment application on the soil solution can also be evaluated by quantifying the concentration of K and NH_4^+ of the soil solution for all control and

Table 4 RIP (mmol kg^{-1}), and K and NH_4^+ in soil solution (mmol L^{-1}) of field samples and distribution coefficients (K_d (Cs); L kg^{-1})) of laboratory samples

Soils amendments	Appl. doses (%)	Initial RIP	Mixture RIP	K	NH_4^+	Initial K_d	Mixture K_d
Sawichi							
Control	–	400	–	1.2	0.8	45	–
Turf	1.4	65	460	0.9	0.4	36	45
	3.0	–	390	0.9	0.6	–	47
Sapropel	1.3	332	400	0.9	0.7	116	40
	2.9	–	360	1.1	0.6	–	39
Dublin							
Control	–	161	–	0.9	0.5	40	–
Mineral soil	17	15	119	1.2	0.6	10	29
	33	–	121	1.3	0.6	–	21
VIUA							
Control	–	24	–	1.0	2.0	25	–
Phosphorite	1.0	810	29	0.6	–	73	26
	3.1	–	20	0.5	1.5	–	26
Polygorskite	0.5	7700	23	0.7	–	376	26
	1.4	–	17	0.5	0.4	–	29
Rudnuy							
Control	–	130	–	0.6	1.7	99	–
Phosphorite	1.4	810	210	0.3	2.2	173	104
	4.2	–	250	0.4	2.0	–	104
Polygorskite	0.7	7700	180	0.4	2.0	902	101
	1.9	–	200	0.3	1.9	–	133
Mateyki							
Control	–	207	–	0.06	0.18	577	–
Sandy soil	6.3	31	170	0.03	0.18	137	546
	12.6	–	250	0.05	0.17	–	523
Loamy soil	6.3	455	260	0.04	0.20	–	–
	12.6	–	320	0.05	0.15	–	–

treated sites (see Table 4). Although a high variability was observed for field samples, the addition of the amendments did not lead to a significant variation in potassium concentrations in the amended soils. Considering the low *potassium* concentrations in some of the control plots (see VIUA, Rudnuy and Mateyki sites), it would have been interesting to increase the fertilizer rate along with the use of the amendments as for this range of K concentration (around 0.5–1 mM), an increase in the K concentration in the soil solution would lead to a decrease in radiocaesium root uptake (Smolders et al. 1997a, b). A significant decrease in the NH_4^+ concentration occurred at the VIUA site when amended with polygorskite. This outcome suggested a beneficial secondary effect when using this amendment, explaining the

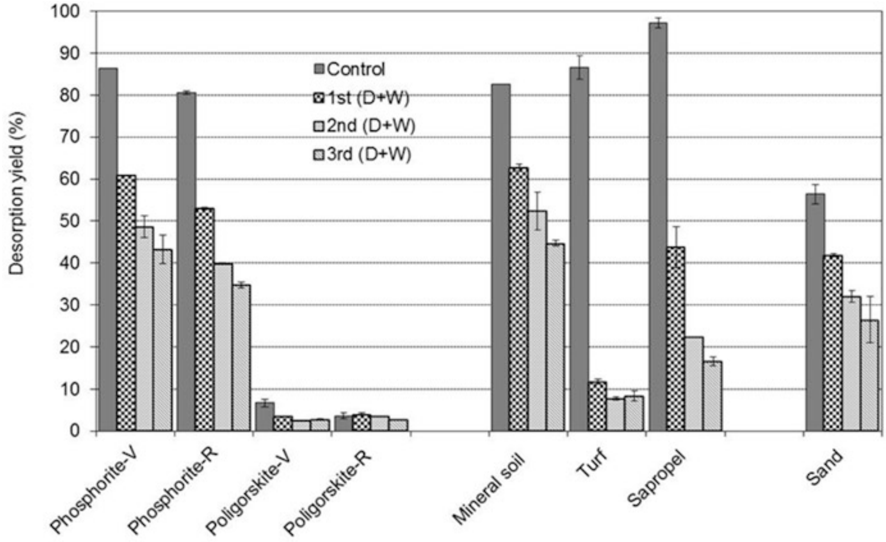

Fig. 3 Variation of radiocaesium desorption yields in amendments exposed to 1, 2 and 3 drying-wetting (D+W) cycles. *Error bars* indicate one standard deviation

decrease in the radiocaesium transfer to plants at the VIUA site when using polygorskite, since both RIP and K^+ concentrations remained constant.

As a way to further characterize the materials (and as a suggested strategy to previously characterize materials at laboratory level before being applied at field). Table 4 summarizes the K_d values obtained for soils, amendments and mixture in laboratory conditions, in which the K_d was calculated in an ionic media representative for each soil solution. Only in the case of polygorskite clay at the Rudnuy sites the K_d (Cs) was one order of magnitude higher than in the soil. In a few cases, the amendments had a much lower sorption capacity than that of the corresponding soil, as was the case of the mineral soil at the Dublin site and the sandy soil at the Mateyki site. The K_d of the soil + amendment mixtures confirmed the lack of the suitability of the materials and administration rates tested. Promising materials, such as polygorskite, were used at such low doses that their effect was diminished. Other materials, such as the mineral soil at the Dublin site, were used in higher doses, but their low K_d led to a decrease in the K_d of the mixture with respect to the initial soil.

Additionally, there may be a significant decrease in the reversibility of the Cs sorption in the medium and long term, which is responsible for a decrease in the Cs root uptake with time and that must be considered for a better assessment of the eventual amendment performance (Absalom et al. 1999; Rigol et al. 1999b). Figure 3 shows the variation of radiocaesium extraction yields in amendment samples exposed to drying-wetting cycles, which has previously been shown to be a good laboratory approach to simulate dynamics at field level (Rigol et al. 1999b). The initial extraction yields ranged from 3.6 % in the polygorskite, to 97 % in the sapropel. Therefore, initial extraction yields were generally high in all the amendments tested, showing that the initial sorption was almost reversible, with the exception of the polygorskite clay, which showed an extremely low radiocaesium extraction yield. Application of such cycles leads to decreases in the extraction yields proportional to the number of cycles applied, indicating that the amount of radiocaesium fixed by the amendment could increase with time. Therefore, interaction dynamics were significant for all samples; with a decrease in the extraction yields of up to one order of magnitude (see the turf material). Again, the polygorskite clay was an exception to this pattern as its final extraction yield was only slightly lower than the initial low yield.

2.6 Prediction of the Effect of the Materials from Sorption and Desorption Data

The extent of the effect of the addition of amendments, in terms of changes in the sorption-desorption pattern of the soil, can be rationalized based not only on the changes in the RIP in the soil-amendment mixture, but also by correcting the fraction of radiocaesium reversibly sorbed at a given time. As stated before, changes in the radionuclide sorption-desorption pattern in the soil will be the predominant effect affecting root uptake, considering that only small changes in the exchangeable complex and in the soil solution can be anticipated due to the low amendment doses used. The equation for a numerical estimation may be written as follows for the case of radiocaesium:

$$Effect = \frac{RIP_s \cdot \left(1 - w_{amend}\right) / f_{rev,s} + RIP_{amend} \cdot w_{amend} / f_{rev,amend}}{RIP_s / f_{rev,s}},$$

in which RIP_s refers to the radiocaesium sorption potential of the soil, $\mathrm{RIP}_{\mathrm{amend}}$ refers to the radiocaesium sorption potential of the amendment, $\mathrm{w}_{\mathrm{amend}}$ is the dose in grams of amendment per gram of soil-amendment mixture, $\mathrm{f}_{\mathrm{rev,s}}$ is the fraction of radionuclide reversibly sorbed in the soil, and $\mathrm{f}_{\mathrm{rev,amend}}$ is the fraction of radionuclide reversibly sorbed in the amendment. This equation is time dependent since radiocaesium reversibly sorbed fraction will significantly change with time. This equation is useful to calculate the amendment dose to achieve a given effect, or to predict an effect with an affordable administration rate, besides also considering significant secondary effects that may affect radionuclide root uptake and eventual concentration in plants (such as changes in the exchangeable complex and in the soil solution composition and plant biomass). Besides, the term RIP can be substituted by the K_d (*Cs*), if the radiocaesium distribution coefficient is calculated at the same soil solution scenario for both the soil and the amendment. Unless the sorption capacity of the materials is at least 2–3 orders of magnitude higher than that of the untreated soils no effects will be observed in Cs mobility (such as root uptake), considering the low administration rate (often lower than 5 %) that can be applied at field level. From the materials tested in the study described above only polygorskite could be considered as an adequate material, although the low administration rate (<1.9 %) made it difficult to observe an unequivocal beneficial effect on reducing Cs transfer.

3 Remedial Options for Agriculture

The ingestion pathway dominates in many contamination scenarios. Therefore, the application of remedial options to intensively and extensively farmed areas is a critical part of many remediation strategies. Subsequently, remedial options need to be targeted on various media (soil, plants, and water) and contamination pathways from the media to crops. The remedial options for agricultural systems should not only be aimed at addressing dose concerns, but also a wide range of other issues such as maintaining the local economy, promoting/upholding consumer trust and ensuring appropriate disposal of wastes (Howard 2012). The soil crop pathway includes (1) arable land used for the production of crops intended for the human food chain (including cereals, vegetables and horticultural crops, and fruit) and for non-food crops for industry (such as flax, bioenergy or biofuel crops) and (2) grassland[6] used for the production of fodder crops such as hay and silage intended as feed for animal consumption. The various available remedial options that intervene along the soil-crop pathways can be grouped according to two main tasks, namely:

- removing or burying most of the contamination, which also reduces external and potential inhalation doses to workers and
- reduction of the soil-to-plant transfer of radionuclides, through a variety of techniques including forms of ploughing, crop selection and soil treatments.

3.1 Selection of Crops and Varieties with Low Accumulation of Radiocaesium

Radiocaesium transfer to plants is highly affected by its physical and chemical properties, soil properties, and the characteristics of different plant species and varieties (Table 5) (Sanzharova et al. 1996). The selection of crops and/or varieties with lower accumulation of radiocaesium is a remedial option is relatively easy to implement and acceptable to farmers as it requires little additional cost and can be implemented within routine farming practice. The decision on which crop or variety to grow should take into account the yield that would be obtained and the relative difference in radiocaesium accumulation between the former and new crop. The main

[6] The term grassland is used here to refer to both cultivated and uncultivated land used as either pasture for grazing animals or for growing fodder. Pasture is used in the report when referring to land used for grazing animals. The term meadow is used in the report for case studies in the FSU and refers to uncultivated grassland used for grazing animals or to grow some fodder crops.

Table 5 Radiocaesium transfer factors for different plant species, based on IAEA TECDOC 1616 (Adopted from Sanzharova et al. 1996)

Plant group	Plant compartment	N	Mean	Min	Max
Cereals	Grain	470	2.9×10^{-2}	2.0×10^{-4}	9.0×10^{-1}
	Stems and shoots	130	1.5×10^{-1}	4.3×10^{-3}	3.7
Maize	Grain	67	3.3×10^{-2}	3.0×10^{-3}	2.6×10^{-1}
	Stems and shoots	101	7.3×10^{-2}	3.0×10^{-3}	4.9×10^{-1}
Leafy Vegetables	Leaves	290	6.0×10^{-2}	3.0×10^{-4}	9.8×10^{-1}
Non-leafy Vegetables	Fruits, heads, berries, buds	38	2.1×10^{-2}	7.0×10^{-4}	7.3×10^{-1}
Leguminous Vegetables	Seeds and pods	126	4.0×10^{-2}	1.0×10^{-3}	7.1×10^{-1}
Root crops	Roots	81	4.2×10^{-2}	1.0×10^{-3}	8.8×10^{-1}
	Leaves	12	3.5×10^{-2}	6.0×10^{-3}	4.5×10^{-1}
Tubers	Tubers	138	5.6×10^{-2}	4.0×10^{-3}	6.0×10^{-1}
Grasses	Stems and shoots	64	6.3×10^{-2}	4.8×10^{-3}	9.9×10^{-1}
Fodder Leguminous	Stems and shoots	85	1.6×10^{-1}	1.0×10^{-2}	1.8
Pasture	Stems and shoots	401	2.5×10^{-1}	1.0×10^{-2}	5.0
Herbs	Stems, leaves	4	6.6×10^{-2}	4.8×10^{-3}	2.8

Table 6 Variety-specific differences in ^{137}Cs accumulation in different agricultural plants (Adopted from Alexakhin 1993)

Plants	No of varieties studied	Range of transfer factor (B_v) values specific for varieties, dimensionless	Ratio of maximum B_v to minimum B_v
Spring wheat, grain	15	0.015–0.046	3.1
Winter rye, grain	11	0.015–0.068	4.5
Barley, grain	11	0.013–0.041	3.2
Oats, grain	4	0.018–0.072	4.0
Potato, tubers	5	0.029–0.058	2.0
Beetroots, roots	4	0.065–0.089	1.4
Tomato, fruit	5	0.01–0.029	2.9
Lupine	16	0.032-0.112	3.5
Clover	8	0.016–0.057	3.5
Vetch	8	0.023–0.065	2.8

reasons for the plant related variations in radiocaesium transfer factors is that crop differ in many factors such as their requirement for certain nutrients such as K, P etc.; in detoxification and exclusion mechanisms; specific patterns of distribution of roots in the soil and rhizosphere properties such as the presence of mycorrhiza (Smolders and Merckx 1993).

Crops selection for cultivation using normal agricultural practices needs to take into account the deposition density of soils and reported F_v values for different crops/cultivars. For example, crops with high F_v values for the type of soil are not grown in areas with a high deposition density of radiocaesium. Evaluation of the major varieties of the farm plants in areas affected by the Chernobyl accident demonstrates more than tenfold inter-crop differences and up to 4.5-fold for inter-variety differences (Table 6). When new varieties of crops are included into an established crop rotation there may need to be a reassessment of fertilisation schemes due to a modification of nutrient requirements. Evaluation of relevant information and dialogue with farmers or other operators will help to identify which crop variety substitutions are appropriate and ensure suitable modified agricultural practices are adopted. A market for alternative crop varieties should exist or would need to be created. Differences in accumulation of radiocaesium by different varieties provides a possibility for selection of plants and plant cultivars with low accumulation of radionuclides to be used in the contaminated areas if other requirements are met such as productivity, resistance to insects and plant susceptibility to diseases. The effectiveness of this approach, in terms of the crop contamination reduction, can vary on average from five- to tenfold, but could be higher if new varieties with low accumulation of radiocaesium are identified.

3.2 Ploughing and Soil Removal

There four different ploughing options that can be used for remediation of the agricultural lands contaminated with radiocaesium. Ordinary ploughing and rotary cultivation or disking are part of routine agricultural practice and can be used in arable lands annually or as a part of amelioration (radical improvement) of meadows or pastures. In the latter case, the use of land is restricted for one or two years until the grass has re-established. Other options considered in this section are beyond normal farming operations and require specialised ploughing equipment to be delivered to the remediation sites. All ploughing options are based on mechanical redistribution of radionuclides in the soil profile through dilution or removal of radiocaesium from the top soil. These options can reduce both external and internal exposures. External exposure is reduced because the more contaminated deeper layers are placed lower in the soil profile and the upper less contaminated soil layer serves as a shielding media against irradiation. Internal exposure is reduced because the transfer of radiocaesium from soil to plant is lowered as the plant roots are in less contaminated soil (Table 7). To be effective, ploughing should only to be applied once and should not be carried out again because the buried radionuclides can then be placed higher up the soil column and in the rooting zone once more.

An ordinary single-furrow mouldboard plough can be used to mix radiocaesium in the top 18–30 cm of the soil profile. Much of the contamination at the surface will be buried more deeply in the soil thereby reducing plant uptake by up to threefold, with an average reduction factor of 2.0. External dose may be reduced from two- to fivefold, depending on the depth of ploughing (RIARAE 1987, 1988). An average reduction factor for root uptake of three- to fivefold may be achieved from deep ploughing with a maximum reduction factor of 10 (Alexakhin 1993;

Table 7 Reduction factors for different types of ploughing options implemented for remediation after the Chernobyl accident (Adopted from Alexakhin 1993; Bogdevich 2002; Fesenko et al. 2007)

Remedial options	Ploughing depth, cm	^{137}Cs average reduction factor in plants	Reduction factor of external dose rate
Soil removal	5	10–20	10–100
Ordinary ploughing (first application)	18–25	2.0 (1.5–3)	2–4
Deep ploughing	30–50	3.0 (2.0–7.0)	2.0–20.0
Skim and burial ploughing	50–70	>20	>20
Rotary cultivation, disking (first application)	10	1.7 (1.5–2.0)	1.0–1.5

Bogdevich 2002; Fesenko et al. 2007; Maubert et al. 1993; Vovk et al. 1993). The external dose may be reduced by 2–20 folds with the highest reduction factors achieved for complete inversion of soil (Alexakhin 1993; Bogdevich 2002; Fesenko et al. 2007). The measure would not be suitable in regions with thin topsoils as soil fertility and structure would be detrimentally affected. There may be resistance to topsoil burial because of the associated impact on soil-dwelling and other flora and fauna. Future restriction on deep tilling may be imposed to ensure that the radiocaesium is not returned to the rooting zone although subsequent normal ploughing (to ca. 25 cm depth) will not bring much contamination back to the surface. The effectiveness of rotary cultivation, disking is not as effective as other ploughing options with an average reduction of radiocaesium transfer to plants up to two times (Table 7). The option can only be applied to natural, non-arable soils as part of meadow amelioration techniques.

Although, all these options were used after the Chernobyl accident (Alexakhin 2009) their applications were restricted mainly to the regions with fertile clay soil. The limited application of this option can also be explained by the environmental constraints in its application, since most of the soils in the most contaminated areas have a thin fertile layer of soil. If no plants are present, a specialist plough with two ploughshares can be used for skim and burial ploughing. The first ploughshare skims off a thin layer of contaminated topsoil (ca. 5 cm; but adjustable) and buries it at a depth of about 45 cm. The deeper soil layer (ca. 5–50 cm) is lifted by the second ploughshare and placed at the top without inverting the 5–45 cm horizon. Therefore, much of the contamination at the surface will be buried deeply in the soil profile. This procedure reduces both external exposure and root uptake from the contaminants, the negative effect on soil fertility which occurs in deep ploughing is minimised and resuspension is also reduced. More than a tenfold reduction in the contamination of the upper soil layers may occur, if the skim and burial ploughing is optimised according to the contaminant distribution in the soil. An associated reduction in soil-to-plant transfer of 10 and in external dose of around 20-fold may be achieved (Roed et al. 1996).

Skim and burial ploughs need to be available for this remedial option to be applied at a large scale and this may be a problem in the initial stages of remediation as these ploughs are not normally in widespread use. The side effects are similar to those above for deep ploughing. In areas affected by the Chernobyl accident the option was used on a limited scale because of similar environmental constraints to those outlined for deep ploughing (namely thin top-soils as soil fertility and detrimental effect on soil structure). Ploughing may substantially change the landscape and the soil properties. Soil fertility may also be reduced requiring additional fertilisation if the land is used for crop production. There may also be negative effects on biodiversity, particularly for soil dwelling organisms, and therefore soil functioning such as decomposition rates. Field drainage systems may be destroyed leading to waterlogging. This remedial option may result in resuspension of radionuclides associated with small soil particles during and after ploughing until the formation of a new root mat in the upper soil layer.

The most contaminated top soil layer can be removed using road construction equipment such as graders, bulldozers, front end loaders, excavators and scrapers or a turf harvester. Removal of the upper soil will take away much of the contamination. The depth of the soil layer which can be removed depends on the thickness of the fertile layer of contaminated soil if applied for agricultural soils. If the original depth of fertile soil layer is not greater than that removed then additional fertile soil would need to be added with acceptably low radionuclide activity concentrations (topsoil replacement). The removal of much of the contamination at the surface will greatly reduce (1) radionuclide uptake by plant roots; (2) external exposure, and (3) resuspension of radionuclides from the soil (Alexakhin 2009). Topsoil removal (or replacement) was used as a remedial option in the FSU following the Chernobyl accident, in Spain after Palomares incident, in Brazil after the Goiania incident and most recently and extensively in the areas affected by Fukushima Dai-ichi accident (IAEA 2015).

After top soil removal, the reduction in radiocaesium contamination of the top soil may reach 10–100 folds, whilst that for soil-to-plant transfer may be 10–20 folds (IAEA 2006, 2015; Alexakhin 2009). In some cases it may be advantageous to remove part of the vegetation cover before removing the layer of soil. The efficiency of removal of the surface layer may be affected by the degree of optimisation achieved in a wide range of factors. These include: the thickness of the removed layer, surface unevenness, presence of rock and stones, soil texture and moisture content and vertical radionuclide distribution relative to the depth of the soil removed. Soil removal may generate a large volume of contaminated waste and issues related to disposal of this waste should be carefully planned before remediation starts.

3.3 Arable Soil Fertilization

Application of mineral fertilizers is routinely used in agriculture to enhance crop yields. The fertilizer is normally comprised of nitrogen, phosphate and potassium fertilisers which are mixed into the soil by harrowing or ploughing before planting/

sowing of arable crops. Mineral fertilizer application rates are adjusted according to crop requirements based on the soil properties, recommended crop cultivation technologies and existing farming practice (Alexakhin 1993; Alexakhin et al. 1996; Nisbet 1993). Application of mineral fertilisers as a remedial option involves a change in both the ratio and application rates of the individual elements (i.e. N, P and K) in the NPK mix applied onto contaminated lands. The use of fertilisers to reduce plant root uptake of radiocaesium are based on decreasing the Cs:K ratio in the soil solution whilst maintaining optimal growth conditions for plants (Fesenko et al. 2007). Mineral fertilisers have been used extensively in all three FSU countries to reduce uptake to plants from contaminated soil in areas affected by the Chernobyl accident. The numerous studies performed after the Chernobyl accident has made it possible to identify the optimal ratios of nutrients for land contaminated by radiocaesium in the contaminated areas (Table 8). In particular, because potassium is a chemical analogue for caesium its application in elevated rates was identified as an effective remedial option to reduce the accumulation of radiocaesium in crops. The optimum ratio of minerals to achieve the maximum reduction in root uptake of radiocaesium was determined to be a N:P:K ratio of 1:1.5:2 (RIARAE 1988). The ratios given are the relative amounts used for each element (N, P and K) for remediation compared with that used for normal application rates. Thus, N:P:K of 1:1.5:2 means the same amount as for normal practice is applied for *N*, 1.5-fold higher for P and twofold higher for K.

Table 8 Reduction factor for radiocaesium accumulation in farm crops for different combinations of mineral fertilisers on leached chernozem

Application rate of fertilisers	Winter wheat	Spring wheat	Oats	Barley	Mean
$P_{60}K_{60}$[a]	1.0	1.0	1.0	1.0	1.0
$N_{90}P_{60}$	1.0	1.0	1.0	1.0	1.0
$N_{90}K_{90}$	1.0	1.0	1.0	1.0	1.0
$N_{60}P_{60}K_{60}$[b]	1.06	1.1	0.9	1.0	1.0
$N_{90}P_{60}\hat{E}_{60}$	1	1.1	0.75	1.1	1.0
$N_{120}P_{60}K_{60}$	0.8	0.7	1.0	0.8	0.8
$N_{150}P_{60}K_{60}$	0.7	0.8	0.6	0.6	0.7
$N_{90}P_{30}K_{60}$	1.2	1.2	1.0	1.0	1.1
$N_{90}P_{60}K_{60}$	1.2	1.0	1.2	1.4	1.2
$N_{90}P_{90}K_{60}$	1.2	1.1	1.2	1.3	1.2
$N_{90}P_{120}K_{60}$	1.3	1.4	1.3	1.5	1.4
$N_{90}P_{120}K_{120}$	1.2	1.5	1.3	1.7	1.4
$N_{60}P_{60}K_{90}$	1.5	1.8	1.6	2.3	1.8
$N_{60}P_{60}K_{120}$	1.7	1.9	2.0	2.0	1.9
$N_{60}P_{60}K_{150}$	1.8	2.0	2.0	2.3	2.0
$N_{60}P_{90}K_{120}$[c]	1.9	2.0	2.0	2.0	2.0

[a]Notation of $N_xP_yK_z$ means mineral fertilizer application rate where x kg of active N per ha, y kg of active P per ha, z kg of active K per ha are applied

[b]Application rate under normal conditions

[c]Optimal combination

During crop cultivation on contaminated soils, *N* fertilizer application rates need to take into account the expected yield. Increasing *N* application can increase radiocaesium transfer to plants due to soil acidification and its effect on K_d(Cs) (RIARAE 2006). Ammonium fertilizers should, therefore, be avoided, and N fertilizer should be applied in the form of nitrate (Nisbet 1993). Supplementary Mg fertilisation and liming may be required to maintain an optimal ionic equilibrium in both soil and plants. On acid and weakly acid soils, mineral fertilizers are only applied after liming, since the use of mineral fertilizers alone, especially in acidic forms, may increase the acidity of the soil solution and increase radiocaesium transfer to crops (Fesenko et al. 2012). The maximum effect of mineral fertilisers' application can be achieved on pure soddy podzolic soils. Application of mineral fertilizers as recommended in the first post-Chernobyl recommendations for the long-term period (RIARAE 1988) may reduce radiocaesium transfer to plants by two to fivefolds (Table 9). Enhancing the rate of P fertilizer application within an NPK fertilizer reduces radiocaesium uptake by plants on mineral soils by 1.5–3-fold. Factors influencing the effectiveness of the option for radiocaesium are potassium status of the soil/soil solution (see above) and the type of crops (Fesenko et al. 2012).

Table 9 Summary of data for radiocaesium on the effectiveness of mineral fertilizers with recommended N:P:K ratio (Adopted from Brown et al. 1995)

Soils	Country	Plants	Average reduction of ^{137}Cs transfer to crops
Soddy podzolic	Belarus	Winter rye	2.8
		Barley	1.1
		Oats	1.8
Soddy podzolic	Russia	Barley	1.5
		0ats	1.5
		Winter rye	1.5
		Maize silage	1.7
		Potato	1.8
		Beetroots	2.1
		Vegetable	2.2
Soddy podzolic	Ukraine	Cabbage	1.4
		Carrots	1.4
		Beetroots	2
		Oats	1.1
Sandy soils		Maize silage	1.5
		Beetroots	1.8
		Lupin	1.1
		Pears	1.6
Peaty		Cabbage	4
		Maize silage	5
		Beetroots	3
		Rape	2–3
		Grass	1.5

Although there may be changes in nutrient status etc., the impact is likely to be small for intensively managed arable soil where mineral fertilisers are routinely applied at normal rates. K and P are the key elements which improve soil fertility and, therefore, increases in their application rates often leads to higher crop yields. Assuming that this remedial option is carried out for soils where the exchangeable K is sub-optimal for the crop, there will be a potential increase in crop yield and quality. Application of phosphates may reduce the availability of essential micronutrients which, when complexed with phosphates, are also of low solubility.

3.4 Liming of Arable Soils

Liming of soil is part of conventional agricultural practice for acidic soils which are present in some areas that are most affected by the Chernobyl accident. The effectiveness of liming is mainly based on neutralization of soil solution acidity, displacement of hydrogen ions from the soil sorbing complex and calcium saturation in the exchangeable complex. Thus, application of lime to maintain a neutral pH in the soil solution may substantially reduce radiocaesium transfer to plants since radiocaesium accumulation in plants is enhanced in acidic soil solutions (Nisbet 1993).

Table 10 Summary of data for radiocaesium on the effectiveness of liming with recommended application rates (Adopted from Brown et al. 1995)

Country	Soil	Plants	Reduction in Cs transfer to plants
Belarus	Mineral	Cereals	1.5–3
	Organic	Cereals	1.5–3
Russia	Mineral	Barley	1.9
		Oats	2.1
		Winter rye	1.8
		Potato	2.3
		Maize	1.6
		Beetroot	2.1
Ukraine	Mineral	Maize silage	1.8
		Beetroot	2.0
		Lupin	2.7
		Peas	2.4
	Organic	Cabbage	2
		Maize	1
		Beetroot	1

Based on the post Chernobyl research in the contaminated areas the application rates of lime for these regions was recommended to be increased on average by 1.5 times from the normal application rate. To maintain the effect, the liming should be applied every four-five years with an application rate from 2 to 10 tonnes per ha (Table 10). Liming can be applied in different forms such as: dolomite powder, calcareous tuffs (travertine[7]) and marlstone[8] (Gulyakin et al. 1978; Ratnikov et al. 1992). The amount of lime used depends on pH and other soil properties (CEC, calcium status, granulometric composition, organic content). Lime is normally applied as an ameliorant[9] to soils of a low pH or low Ca status, but the application rate and the frequency of application is determined by the soil fertility (Table 10). It is normally ploughed into the soil before arable crops are planted or sown. When the total amount intended to be applied over a growing season exceeds 8 t ha^{-1} then lime is applied on two occasions: half during ploughing and half during the plant growth period as this has a greater sustained impact on soil fertility (Bogdevich 2002).

In European countries (with higher levels of soil fertility compared with the FSU countries) maintenance liming normally takes place every 5 years (0.5–2 t $CaCO_3$ ha^{-1}; depending on the soil pH) with the aim of reaching pH 7 in mineral soils and pH 6 in organic soils (Woodman and Nisbet 1999). Liming may reduce radiocaesium transfer to farm products by 1.5–4.0 folds (Table 11) respectively depending on factors such as the initial soil pH, CEC and calcium status, hydrological regime of the soil, productivity and type of crops. The effectiveness is usually higher on organic soils than on mineral soils (Fesenko et al. 2012) (Table 10). Liming can change the microbiological status of the soil and may affects water quality. There is a potential secondary negative effect on radiocaesium transfer in soils with a low potassium

Table 11 Summary of data for radiocaesium on the effectiveness of organic fertilizers (Adopted from Brown et al. 1995)

Country	Fertiliser type	Plants	Reduction in plant uptake
Belarus	Manure, 60 t ha^{-1}	Potato	1.5–2.8
	Sapropel, 60 t ha^{-1}	Various crops	1.7
Russia	Manure	Maize silage	2.4
		Barley	1.6
		Oats	1.3
		Winter rye	1.4
		Potato	2.9
		Beetroot	2.2
		Vegetables	2.9
Ukraine	Manure, 50 t ha^{-1}	Maize silage	2
		Beetroots	4
		Lupin	1.3
		Pears	2
	Sapropel, 100 t ha^{-1}	Beetroot	4–5
		Peas	2
		Lupin	3
		Maize silage	2

[7] Travertine is a terrestrial sedimentary rock, formed by the precipitation of carbonate minerals from solution in ground and surface waters.

[8] Marlstone is a calcium carbonate or lim*e*-rich mud or mudstone which contains variable amounts of clays and aragonite

[9] A substance added to soil to improve the growing conditions for plant roots

adsorption ratio (much higher Ca than K concentrations) and low K concentration (lower than 0.5 mM) in the soil solution. In these cases, there may be a partial loading of Ca at the FES, leading to a lower K_d(Cs) values (Fesenko et al. 2012). Therefore, liming should be accompanied by K fertilisation to prevent this process. Liming may also induce microelement deficiencies in crops (in particular, Mn, Zn) and additional application of micro fertilizer[10] may be necessary.

3.5 Application of Organic Materials to Arable Soils

The application of organic materials may reduce radiocaesium transfer to plants by 1.3–5-folds (Alexakhin 1993, 2009; Brown et al. 1995). The effectiveness of organic material fertilizer for radiocaesium accumulation in plants is higher on low fertile light sandy soil compared with loamy soils, due to the large difference in the CEC values (Table 11).

Application of organic materials is a conventional farming practice which should be slightly changed to provide a reduction of radiocaesium transfer to plants. Organic material applied specifically to decrease radionuclide transfer to plants may be of different origins and can include manure, straw and plant derived fertilizers (species such as lupin and serradella) (Belova et al. 2004; RIARAE 2006). Peat and sapropel may also be used as soil ameliorants (RIARAE 2006; Fesenko et al. 2012). Sapropel is formed from bottom sediments in natural lakes and consists of plant and animal residues decomposed in anaerobic conditions. The main advantages of sapropel are a high content of organic matter (up to 70 %), with a high content of humic acids and nitrogen, high CEC, the presence of mineral matter and mobile forms of nutrients (Brown et al. 1995). However, sapropel may be acidic (pH 4.5–6.5) leading to reductions in the pH of soil solution. Depending on the prevailing soil acidity this may lead to some increase in radionuclide transfer to plants (Fesenko et al. 2012).

Organic materials are normally applied to soils with a low organic content and of light granulometric texture. They are easy to apply and increase plant production by enhancing the nutrient and microelement content of treated soils. The conventional application rate of organic material depends on soil properties including the organic content, CEC and granulometric composition as well as the type of crop. Increased application rates of organic fertilisers were used widely in the FSU following the Chernobyl accident on arable land with deposition densities above 185 kBq m^{-2} (Fesenko et al. 2007). The application rates recommended were 1.5–2-folds higher than those used normally (RIARAE 2006; Fesenko et al. 2012). The timing of organic fertilization is crop dependent. For example, peat-manure composts are applied in spring for a planned yield of medium and late cultivars of potato, whereas for early potato it is applied in autumn during winter ploughing. Because organic

[10] Microfertilisers are fertilisers containing microelement, i.e. chemical elements which the plant requires in microquantities.

fertilizers are routinely applied on intensively managed arable soils the side effects are minimal. Crop yield may be increased by up to twofold and an associated improvement in soil fertility can be expected. Application of organic fertilizers such as manure and acid peat may increase the uptake of radiocaesium by plants in the first year after application because of changes of soil acidity. However, in the second year after application, organic fertilizers may produce a decrease in radiocaesium uptake by plants due to mineralization in soil leading to an increase in the content of potassium in the soil solution.

3.6 Application of Mineral Sorbents to Arable Soils

Mineral sorbents used for remediation may have different origins. The most commonly used materials are clays (such as bentonites and palygorskite) and zeolites (such as clinoptilolite), because these materials have a high sorption affinity for certain radionuclides. Application of mineral sorbents is not normally part of normal farming practices in most areas. Mineral sorbents added to soil enhance the sorption capacity of the soil so they should have a much higher sorption capacity for the target radionuclide than that of untreated soils (Sawney 1964; Nishita et al. 1968; Bakunov and Yudintseva 1989). A recommended particle size for mineral sorbents added to soil is 1 mm or lower to maximize sorption capacity. Zeolites can be used as a substitute for lime, due to its high pH, and it can be also added to peat-manure compost. The dry sorbent material should be uniformly spread on the soil surface before planting, and then the soil is re-ploughed. The mass of the applied material depends on the contamination level and soil properties. Application rates vary from 5 to 30 t ha^{-1}, the upper value was recommended for light sandy soil following the Chernobyl accident. When zeolite is applied, increased doses of mineral fertilisers and microelements should also be used because of their additional sorption by the sorbents implemented for remediation.

The use of mineral sorbents may reduce radiocaesium transfer to crops up to 2.5-fold depending on soil texture (RIARAE 2006). The maximum effectiveness may be observed for light sandy soil with low fertility and low K_d values such as sandy soils; the effect is low for clay soils with a high fertility (and high CEC status). The effectiveness of mineral sorbent addition tends to increase with time. Positive effects from the use of zeolites, for instance, appear from the second or third year after application when there has been an adequate amount of time for the clay to form a sorbing complex with radiocaesium in the soil (Wilkins et al. 1996). Mineral sorbents for soils need to be available in large amounts to be applicable at field level. Application of mineral sorbents can change the nutrient status and CEC of soil. The use of sorbents can stabilize the sorption properties of soil and improve its fertility providing positive effects for crop planting. One detrimental aspect of the application of the mineral sorbents as a remedial option is its relatively high cost. If there

Table 12 Summary of the data on effectiveness of combined application of mineral fertilisers and lime on radiocaesium accumulation in plants

Combinations of options	Reduction in radiocaesium accumulation in plants
$N_{60}P_{90}K_{120}$	2.0
$N_{60}P_{90}$K60	1.4
$CaCO_3$ (1Hg)	1.6
$CaCO_3$ (1.5Hg)	1.9
N60$P_{90}K_{60}+CaCO_3$ (1Hg)	1.9
$N_{90}P_{60}K_{90}+CaCO_3$ (1Hg)	2.0
$N_{60}P_{60}K_{90}+CaCO_3$ (1.5Hg)	2.2
$N_{60}P_{90}K_{120}+CaCO_3$ (1Hg)	2.6
$N_{60}P_{60}K_{90}+CaCO_3$ (1.5Hg)	2.5
$N_{60}P_{90}K_{120}+CaCO_3$ (1.5Hg)	3.2

are local deposits of mineral raw materials close to the contaminated areas these costs may be reduced.

3.7 Combination of Soil Based Options

Most of the remedial options discussed earlier can be applied individually and in combination. A combined use of mineral, organic fertilizers and liming is the most effective way to reduce radionuclide accumulation in farm crops. The combined use of lime and organic matter may reduce radiocaesium transfer to plants by up to three- to fivefold and the effects persists into the second and third years after the application. The use of increased application rates of P-K fertilizers combined with liming reduces transfer of radiocaesium by three- to fivefolds respectively. As an example Table 12 provides summary data on effectiveness of combined application of mineral fertilizers and liming.

A combined use of liming and increased rates of P-K fertilizers together with normal application rates of N can decrease radionuclide accumulation in farm crops up to two to fourfolds more than after liming alone. Many crops are sensitive to a deficiency of microelements, therefore, the additional application of micro fertilizer[11] is essential for remedial options such as liming and use of increased doses of P fertilizers.

[11] Microfertilisers are fertilisers containing microelement, i.e. chemical elements which the plant requires in micro quantities.

3.8 Amelioration of Grasslands

Remedial options for meadow soils normally combines some options already mentioned such as disking, ploughing and crop selection with additional procedures which are often used within conventional farming practices. These options involve adapting standard operations to achieve a maximal reduction in radiocaesium transfer to fodder plants in certain site specific conditions. There are two basic techniques of amelioration applied after the Chernobyl accident, namely radical and surface improvement. As contamination of milk and milk products is the main contributor to internal exposure of the affected population, radical improvement of meadows and pastures was a key element of the remediation policy applied in the contaminated regions. As well as reducing radiocaesium transfer to plants, both options have the additional benefit of increasing the productivity of lands used for fodder production and can be used for both uncultivated and cultivated grassland.

Surface improvement includes land improvement (extraction of shrubs, small hillocks of grass, weed control), application of soil mechanical treatment techniques (disking of the root mat which involves disrupting the root mat using heavy discs in 2–3 cuts) and enhanced mineral fertilization. In addition grasses are re-sown in the third year following the commencement of the procedure, thereby improving productivity of plants and ensuring appropriate ratios amongst the different species in the grass mix (RIARAE 1988; Sanzharova et al. 1996; Vidal et al. 2000). The grass mix is includes plant varieties with low accumulation of radiocaesium. The option includes application of lime materials (on acid soils) and increased amounts of P and K within an NPK fertilizer. The approach to select ratios between application rates of NPK fertilisers (or nitrogen, phosphorus and potassium) recommended for remediation of grasslands were similar to that of suggested for arable soils, namely, 1:1.5:2 compared with normal rates of fertilizer application (RIARAE 1988; Sanzharova et al. 1996). The optimal administration rates (active substance per ha) designed to minimise radiocaesium transfer to fodder plants are: (1) $N_{120}P_{90}K_{120}$[12] for dry grassland on mineral soils, (2) $N_{120}P_{90}K_{120}$ for dry grassland on floodplain soils or (3) $N_{180}P_{120}K_{180}$ for wet grassland on floodplain soils. Liming is obligatory for acid soils with application rates which are 1.5–2 folds higher than those normally estimated for the soil solution acidity for fodder land (Sanzharova et al. 1996).

Cereal grasses are preferable on radioactively contaminated lands; since accumulation of radionuclides by cereal grasses is c. two- to threefold lower than that by legume grasses. The grass composition used for surface improvement may include up to 20 % of legume grasses (white clover, vetch). A proportion of land may be sown with perennial grasses which are suitable for both hay production and animals grazing in early spring (in Russia 20 % brome grass was recommendation after the Chernobyl accident (RIARAE 2006). This remedial option was used extensively in the FSU after the Chernobyl accident (Fesenko et al. 2007).

[12] Notation of $N_xP_yK_z$ means mineral fertilizer application rate where x kg of active N per ha, y kg of active P per ha, z kg of active K per ha are applied.

Radical improvement consists of similar options to those of surface improvement with additional soil ploughing. Thus, radical improvement includes removal of shrubs, small hillocks of grass, root mat destruction, ploughing, disking, rototilling and chiselling, liming of acid soil (if necessary), application of increased amounts of P-K within NPK fertilizers and the selection of grass mixtures with the minimum possible accumulation of radionuclides. As above, drainage can also be included for wet soil. The effectiveness of these types of remedial option depends on the state and type of land used for fodder production and varies depending on the type of meadow, the hydrological regime, soil type, nutrient status and pH. The appropriate selection of plant species for reseeding is also important as transfer of radionuclides to different species can vary substantially.

Since the first year after the Chernobyl accident radical improvement has been a key remediation measure carried out extensively in practically all contaminated areas of Belarus, Russia and Ukraine. Radical improvement was effective in field conditions achieving a 3.0–4.0-fold reduction in root uptake of radiocaesium (Table 13). For organic soil the effectiveness increases to 3.0–5.0-fold, with maximum effectiveness (with drainage) of 10–20-fold in wet peat soil. The mechanism of action and effectiveness of radical improvement of meadows strongly depends on the types of meadow and soil properties (Sanzharova et al. 1996; Vidal et al. 2001; Bogdevich 2002). One limitation is that radical improvement (as other agricultural actions involving ploughing) cannot be carried out on sandy soils (due to risk of destruction very shallow fertile layer), steep slopes and river valleys due to the risk of erosion (RIARAE 1991; Brown et al. 1995). Surface improvement is around 20 % cheaper than that of radical improvement. Furthermore, surface improvement may be implemented on erosion-prone sites and low-productivity grassland such as those in floodplain areas where application of radical improvement is restricted. For the case of first application, surface improvement provides reduction of the soil-plant transfer

Table 13 Summary of the reduction factors of different remedial options for grasslands used in the FSU countries (Adopted from Alexakhin 1991, Sanzharova et al. 1996; Bogdevich 2002; RIARAE 1998)

Remedial option	Soils	Reduction of ^{137}Cs transfer to plants
Radical improvement		
First application	Mineral	3.0-4.0
Further applications	Mineral	2.0–3.0
First application	Organic	3.0-20[a]
Further applications	Organic	3.0–8.0
Surface improvement		
First application	Mineral	2.0–3.0
Further applications	Mineral	1.3–2.0
First application	Organic	1.5–6.0
Further applications	Organic	1.5–3.5

For wet peat with drainage

of radiocaesium in a range from 2.0 to 3.0 and 1.5–6.0 for mineral and organic (peat) soils respectively. For further application the effectiveness of this option is about 1.5–2.0 lower. On peaty soils the reduction factor is on average two- to fivefold higher than on mineral soils FSU (Sanzharova et al. 1996). Surface improvement of wet peat soil with drainage (where required) may reduce radiocaesium accumulation in grass by up to a factor of 10 (RIARAE 2006).

Radical improvement may decrease the transfer of radiocaesium to fodder plants by 3–20 folds. Reduction factors for soil-plant transfer of radiocaesium following radical improvement were in the range for mineral soils, from two- to fourfold, and for organic soils from three- to sixfold. A combined option such as drainage and radical improvement of fodder lands may reduce accumulation in grass of radiocaesium by up to a factor of 10. If applied to wet peat soil the reduction factor is greater at up to 20-fold. The option remains effective over 3–5 years and should then be reapplied although it will then have a lower effectiveness. In repeated radical improvement the reduction factors for transfer to plants are 2.0–3.0.

The above options can have potentially high environmental side effects because of the change of ecosystem from natural to cultivated grassland. Disking, application of lime and fertilisers and reseeding will change the ecological characteristics of the land with possible reductions in biodiversity. Grasslands are often the habitat of endangered species and a change in nutrient status may be harmful to these species. A significant increase in NPK application can lead to pollution of ground and surface waters by these elements. When applied on floodplain grassland, water body contamination by fertilizers may occur. Higher productivity of grassland should be anticipated since surface improvement of haylands and pastures increases their productivity by 25–50 % at a minimal cost which can be recovered within 1–2 years. If improvement is carried out under a rolling programme there should be no significant loss of grazing. There may be disruption to farming and other related activities, although there are also benefits to the farmer who will have more improved pastures in the long term. The availability of additional improved grazing can reduce wintering costs and result in higher prices for improved stock. The application of amelioration of meadow is only possible in areas where the soil structure and landscape are suitable. The option cannot be applied on grassland located on waterlogged soils, or on sites where the upper organic horizon is less than 10 cm deep. In the Chernobyl affected areas surface improvement is only applied to dry grassland on soddy-podzolic soils if the resulting plant species stand contains at least 50–60 % of valuable fodder comprising of cereal grasses and 25–30 % of legumes grasses.

3.9 Change in Land Use and in Crop Composition Grown on Contaminated Land

Different types of the land use are associated with different planting regimes. As demonstrated in Section 3.1 (see Table 5), the difference in radiocaesium accumulation by plants can be as great as a factor of 100 dependent on soil and plant properties.

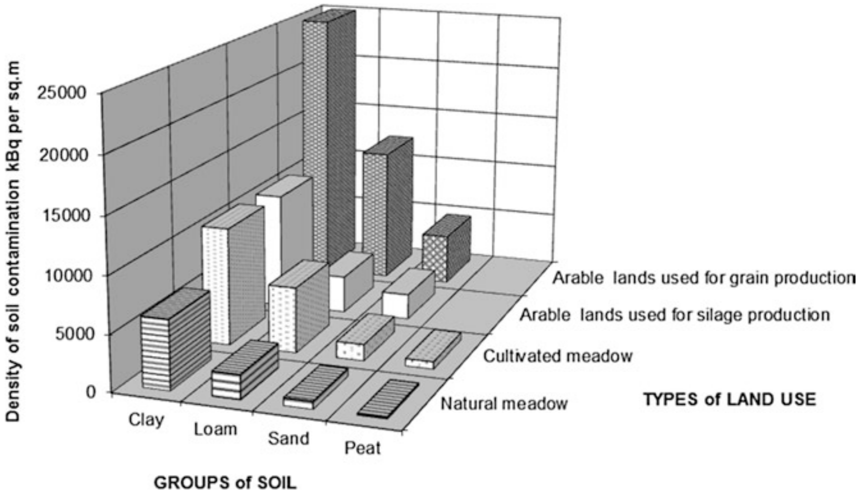

Fig. 4 Densities of soil contamination by ^{137}Cs which would result in exceeding the Chernobyl related temporary permissible (or action) levels (TPLs) for 1994 in various products produced in Russia for different types of land use and soil groups (Adopted from Fesenko et al. 2000a, b)

Therefore, the contamination of agricultural products is specific to every land use, and depends on the soil type and agricultural products produced on contaminated lands. Exclusion of agricultural products vulnerable to the contamination may be an effective method to retain the economic sustainability of affected regions. In particular, contaminated lands may be used for crop production instead of grazing animals or even for non-food produce, such as cotton/flax for fibre; rapeseed for bio-diesel; sugar beet for bio-ethanol; perennial grasses or coppice for biofuel. Agricultural land may also be used for the production of leather and wool. In situations where the land is highly contaminated it may be used for forestry or the placement of suitable industrial enterprises. Such land-use related variation needs to be considered when selecting a possible alternate land use for contaminated regions, together with any variation in permissible levels (TPL) for different agricultural products.

To demonstrate the effect of different land use options Fesenko et al. (2000a, b) used data from the monitoring programme to estimate the ^{137}Cs deposition of soil contamination which would result in exceeding TPLs in various products from different types of land use in 1994 (Fig. 4). There was a marked difference in the amount of ^{137}Cs deposition in soil which would limit the suitability of different types of land use. Using such analyses it was possible to decide whether areas with a defined land use and soil types require remediation, or whether a change in land use might be appropriate. Options such as the conversion of arable land into meadow, converting agricultural land to forestry were implemented in some of the most contaminated areas.

In Belarus, a land use change to rape seed production was applied in contaminated areas with the aim of producing two products: edible oil and protein cake as an animal fodder (Bogdevich 2002). The varieties of rape seed grown had a two- to threefold lower ^{137}Cs and ^{90}Sr uptake rate than many other varieties. Additional fertilisers (Liming 6 t ha^{-1} and fertilization with $N_{90}P_{90}K_{180}$) are used to reduce radiocaesium and radiostrontium uptake into the plant by a factor of about two. This reduces contamination of the seed, which is used for the protein cake. During processing of the rapeseed, radiocaesium is effectively removed, and negligible amounts remain in the final product. The production of rapeseed oil in this way has proved to be an effective, economically viable way to use contaminated land and is profitable for both the farmer and processing industry. From 1992 to 2002 the area under rape seed cultivation in Belarus has increased fourfold to 22000 ha (Bogdevich 2002). The option was widely used in the areas most affected after the Chernobyl accident and was proven to be an effective way to remediate contaminated areas.

4 Remedial Options for Freshwater Ecosystems

Freshwater ecosystems are rather diverse environment and include lakes, rivers and groundwater. Each of these water bodies is unique in terms of the factors governing flows into, within and out of the water body. The effectiveness of remedial options for freshwater ecosystems is dependent on many parameters which are highly site specific and which can substantially constrain potential remediation options that can be applied. The doses arising from freshwater pathways are normally lower than that from terrestrial food. In terms of collective doses to humans, the irrigation and water supply pathways represent the most important pathways. Nevertheless, freshwater-related ingestion pathways may be important contributors to individual doses for people living in settlements surrounding closed lakes, which do not have an outflow. For example, the Kozhany settlement in the Bryansk region, Russia, is located on the shore of a closed lake in an area with peaty soil that was highly contaminated after the Chernobyl accident in 1986. The ^{137}Cs activity concentrations in lake water and fish were two orders of magnitude higher than that from local rivers and open lakes, and remain relatively high for 10–20 years after the accident Travnikova et al. (2004) at 10–20 kBq kg^{-1} fw of ^{137}Cs in lake fish which and exceeded the temporary Russian permissible levels for the fish by a factor of 20–40. The consumption of the fish was up to 50 % of the internal doses to residents of the settlement (Travnikova et al. 2004).

Exposure of the public from contaminated bottom sediments or from sand on the beach can also can occur in highly contaminated areas. However due to the self-shielding of water, external doses from recreational use of contaminated lakes and rivers are normally relatively low (Fesenko et al. 2012). Thus, the focus of implementation of remedial options for aquatic environments is reduction of radiocaesium transfer from contaminated watersheds to water bodies and various options to decrease contamination of edible freshwater species. Relevant remedial options for freshwater can be divided into two main groups: (1) preventing contamination of water bodies by the use of physical or chemical barriers, and/or (2) treatment of water (blending and/or purification). These options are more effective if combined with dietary advice, the provision of monitoring kits, food labelling and compensation schemes (Fesenko et al. 2012).

The first option involves the addition of these barriers to the boundaries of water bodies and therefore can have some features in common with chemical additives used in the second option. Dredging of canal-bed traps to intercept suspended particles in contaminated rivers was carried out after the Chernobyl accident (Voitsekhovitch et al. 1988). These canal-bed traps were highly inefficient for two reasons: (1) flow rates were too high to trap small suspended particles which had adsorbed much of the radioactivity; (2) a significant proportion of the radioactivity (and most of the "available" activity) was in dissolved forms and could not have been intercepted by sediment traps.

4.1 Prevention of Water Body Contamination

The concept behind this option is to prevent remobilisation of runoff radiocaesium from the catchment by installing dykes thereby decreasing long-term transfer to freshwater ecosystems (Smith et al. 2001; Onishi et al. 2007). This measure was widely used after the Chernobyl accident mainly around the Pripyat river floodplain to prevent the secondary contamination of freshwater ecosystems from highly contaminated areas surrounding the river and floodplain. The dyke led to a substantial reduction of radiocaesium transfer to downstream of the Kiev Reservoir and Dnieper River after Chernobyl (Onishi et al. 2007). However, the effectiveness of the option was low for large scale applications since only a small fraction of the total amount of radioactive material deposited in aquatic ecosystems and their catchments retained.

Zeolite containing dykes were constructed on smaller rivers and streams around Chernobyl to intercept dissolved radionuclides. These were ineffective since only 5–10 % of ^{137}Cs in the small rivers and streams were adsorbed (Voitsekhovitch et al. 1997). Buffer strips around rivers can be used to retain radiocaesium contained in eroded particles or in dissolved form and encourage its infiltration and sorption in the soil. This option has been in widespread use within different environment protection schemes to reduce runoff of fertilisers and pesticides from agricultural land to the water (Smith et al. 2001). Removal of radioactivity from water by "filtering" through reed beds is also possible, though probably not feasible on a large scale.

4.2 Fertilization of Water Bodies

The fertilisation of lakes can only be applied to low productivity lakes with low potassium and phosphorus concentrations in the water and low pH. As for agricultural systems, the uptake of radiocaesium by freshwater species from water may be affected by competition with potassium ions, water pH and the presence of other microelements such as phosphorus in the water. Studies on both weapons-test-derived and Chernobyl radiocaesium demonstrates that the uptake to fish (or the concentration factor (*CF*) of radiocaesium in fish) is inversely proportional to the potassium (Fleishman 1973; Blaylock 1982; Smith et al. 2000, 2003). An empirical model for the prediction of radiocaesium concentration factor (CF) values in fish (Rowan and Rasmussen 1994) in the form:

$$CF = \frac{A}{\left[K^{+}\right]^{B}},$$

where CF is measured in l kg^{-1} and $[K^+]$ is in mg L^{-1}, *A* and *B* are the model constants.

The approach was tested against measurements made between 1992 and 1997 in 10 lakes in Russia, Belarus and Ukraine. The model validations, based on measurements of K^+ and suspended solids concentrations in the lake water, were in good agreement with measured values. Therefore, addition of potassium based materials may have the potential to be effectively used to reduce radiocaesium concentrations in fish and other freshwater species. The effectiveness of potassium application strongly depends on the water chemistry at the time of application (e.g. potassium concentration, pH and total phosphorus concentration), the amount and type of K applied and the water retention time. For lakes with rapid inflows and outflows of water, many repeated applications would be necessary to achieve a consistent stable effect (Fesenko et al. 2012). A low effectiveness of this option with a reduction of ^{137}Cs in perch of only 1.1-fold was reported for Swedish lakes affected by the Chernobyl accident (Håkanson and Andersson 1992). Addition of potassium chloride to Lake Svyatoe, Belarus (a lake with a very low natural potassium content), was much more efficient with a tenfold increase in the potassium content of the lake water giving a two– threefold reduction in radiocaesium activity concentration in fish (Smith et al. 2001). For lakes, the effectiveness of potassium application strongly depends on the initial calcium concentration, pH, total phosphorus concentration in the water, amount and type of liming applied and the water retention time. Some modelling assessments have shown that a reduction of about 1.3–1.7-fold in fish can theoretically be obtained (Smith et al. 2001). There may be negative side-effects of the application of various elements to water bodies. Application of mixed K, NH_4, and phosphorus fertilizer may cause eutrophication whilst potassium applied as KOH may increase the pH in lake water. The potential importance of these possible impacts has not been assessed and may be minor in some cases. In one experiment the application of potassium led to a threefold increase in radiocaesium activity concentration in water due to competition with K in sediments (Smith et al. 2000). This side effect would make the option unacceptable if the water is to be used for drinking or irrigation.

Fertilization of lakes with lime and/or phosphorous may be useful as a remedial option for a variety of toxic contaminants in lakes with high productivity, where transfer rates of radionuclides, other metals or organic pollutants may be quite high. Different methods of fertilisation can be used to reduce transfer to freshwater species. Lime or lime mixed with phosphorus can be directly added to the water (or to the ice over the winter period) or used for fertilisation of the catchment of affected water bodies, providing a more gradual source of these substances to the lake water. Different commercial fertilisers as well as phosphorus containing effluents from fish farms can be also used for such fertilisation.

For lakes, effective remediation would need to considerably raise the Ca level, pH and alkalinity and sustain the elevated levels for a long period of time; however, the duration of water chemical responses may be relatively short, depending on the water residence time of the lake. Catchment application may avoid a "liming spike" and considerably prolong the duration of an effect. There is a considerable experience in application of lime in relation to acidification but not for radionuclides. Although, this measure was tested in Scandinavian countries after the Chernobyl accident (Håkanson and Andersson 1992; Outola and Rask 2011), the measure has not been implemented extensively as a remedial option. The effectiveness of potassium application strongly depends on the initial *Ca* concentration, pH and P concentration in the water, amount and type of liming applied and the water retention time. Some assessments have shown that a reduction of about 1.3–1.7-fold in fish can be obtained (Smith et al. 2001). However, in Swedish lakes contaminated after the Chernobyl accident, a low reduction factor of 1.1 was reported for perch fry (Håkanson and Andersson 1992). One negative possible side effect is that liming of naturally acid systems can lead to profound, structural changes in the ecosystem. As a result some changes in the wetland flora may be expected such as the replacement of bog moss by leaf mosses and sedges and damage to sensitive species (e.g. lichens) due to wind drift of lime to adjacent areas (especially if using helicopters) (Smith et al. 2001). The buffering capacity of lakes can be temporarily improved with liming, which has a stabilizing effect on a lake, and is therefore beneficial for lakes (and food chains) that are susceptible to acidification. Catchment liming may also lead to improved conditions for animals and plants in streams and rivers due to reduced transport of metals such as *Fe* and *Al* into the lake from the catchment area (Smith et al. 2001).

5 Forest Ecosystems

Forests are extensive natural resources which provide economic, nutritional, recreational and social benefits to people in many countries so any restrictions applied to the forest due to contamination may result in high economic losses and have severe social and psychological consequences. In many areas subjected to radioactive contamination, forests and forest products were, and still are, an important source of internal and external exposure of the population (IAEA 2006). For example, in the initial years after the Chernobyl accident, the contribution of agricultural products (mainly milk) to the internal dose was much higher than that of forest products. However, the contributions of forest exposure pathways increased with time (Tikhomirov and Shcheglov 1994) from 10 to 15 % in 1987 to

40-45 % in 1996 (Fesenko et al. 2000a, b). Unfortunately, there are only a few practical options that can be implemented in forest ecosystems. The imposition of restrictions on access to forests and consumption of forest products may be effective for a few years but is unlikely to be sustainable in the long term. Therefore, the provision of advice about possible doses from the forest on which products are likely to be contaminated combined with access to monitoring stations to measure contamination is probably a more practically realistic way to reduce such doses (Fesenko et al. 2012).

5.1 Forest Soil Treatment with Fertilizer or Liming

As for agricultural soil, surface application of fertilizers (NPK or PK) or liming in case of acid forest soils in the forest can reduce root uptake of radiocaesium by forest plants and enhance their growth (Fesenko et al. 2000a, b). Such options were tested in Belarus, Russia and Nordic countries after the Chernobyl accidents, but were not used on a large scale (Ipatyev et al. 1999) (see Fig. 5). A good example of the effect of fertilisation with potassium of the forest soils is presented in Rosén et al. (2011) who studied the long-term effects of a single application of potassium fertilizer (100 kg K ha^{-1}) on ^{137}Cs uptake in some understory and fungal species (Figs. 6). The uptake of ^{137}Cs by plants and fungi growing on K-fertilized plots 17 years after application was significantly lower than in similar species collected at non-fertilized sites. The ^{137}Cs activity concentration was 21–58 % lower in fungal sporocarps and 40–61 % lower in plants in the K-fertilized area compared with the control sites. The reductions in ^{137}Cs activity concentration in both understory species and mushroom fruiting bodies were statistically significant for most studied years. The data demonstrates that application of K fertilizer to forests might be effective long-term measure to decrease contamination of plants and fungi, although this option is expensive and cannot be applied in a large scale.

The application of dolomite powder leads to a rise in Ca^{+2} concentrations in soil solution. Bivalent Ca^{+2} cations are not capable of penetrating inward to the wedge-shaped zone of clay minerals, but can stabilize it in a dilated state, during which more ^{137}Cs cations can occupy FES sites. If there is a decline in Ca^{+2} concentration in soil solution, due to vertical migration and other processes in soil, layer collapse of clay minerals ("delayed collapse") is intensified and, consequently, ^{137}Cs fixation is increased in the crystal lattice. Typically reduction factors of 1.5–2 have been achieved by application of dolomite powder in forests, however, the effectiveness is dependent on forest soil fertility and the type of contaminated forest (Ipatyev et al. 1999; Spiridonov et al. 2005). Fertilizer application to the forest soil is generally more efficient if applied in boreal forest than in Mediterranean forest, and fairly efficient in deciduous forest (Ipatyev et al. 1999).

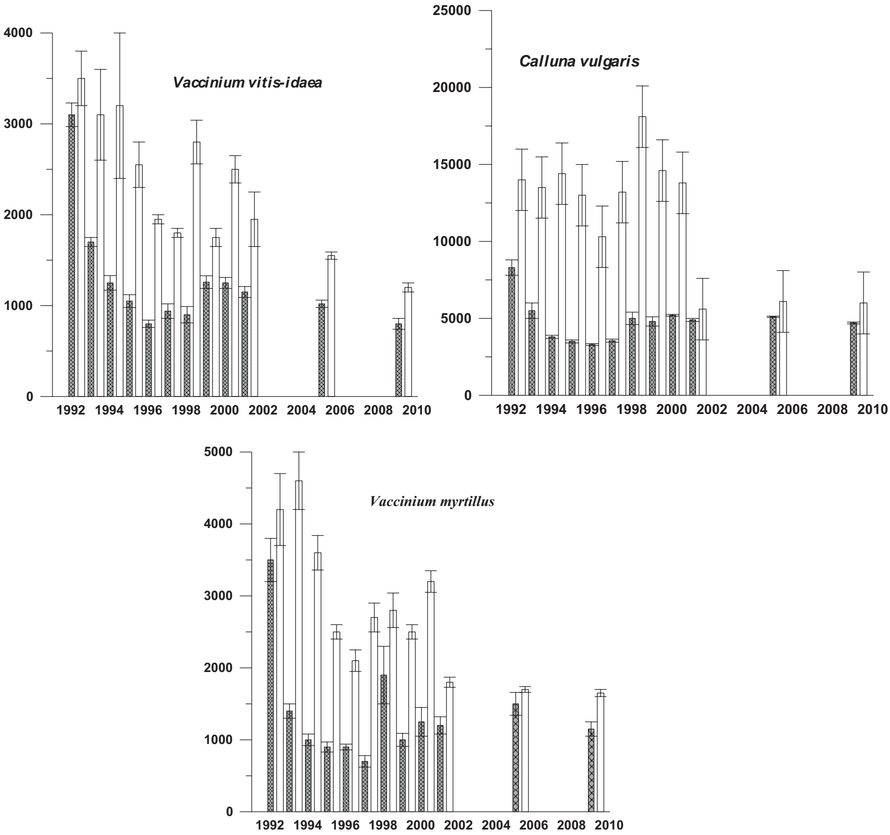

Fig. 5 ^{137}Cs activity concentrations (Bq kg^{-1} d.w.) in heather (*Calluna vulgaris*), bilberry (*Vaccinium myrtillus*) and lingonberry (*Vaccinium vitis-idaea*) in potassium treated and control plots in 1992–2009 (Adopted from Rosén et al. 2011)

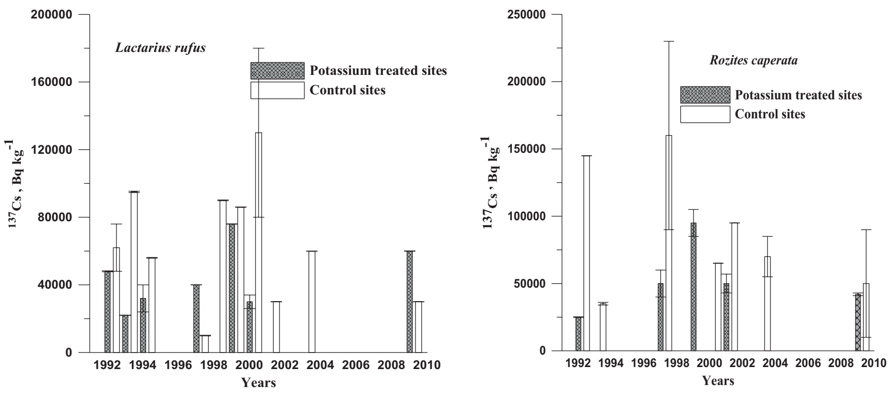

Fig. 6 ^{137}Cs activity concentrations (Bq kg^{-1} d.w.) in some fungal species in potassium treated and control plots in *Lactarius rufus* and *Rozites caperata*

5.2 Modification of Tree Felling Schedules

The remedial option is based on changes in the timing of tree harvesting, intermediate felling and other forest services with the aim to minimize root uptake of radiocaesium in stem wood, or to delay wood harvesting so that processes such as soil immobilisation, vertical migration down the soil profile or physical decay of radionuclides reduce the contamination in the wood. Early felling is most suitable for mature or nearly mature trees soon after the time of contamination whilst delayed felling is most suitable for young trees reaching maturity about 20 years after contamination (Fesenko et al. 2012). The effectiveness depends on factors such as which radionuclides dominate in the long term, their radiocaesium activity concentrations in the forest, and forest characteristics such as age of trees, productivity, forest soil, type of understory (Frissel et al. 1996). Overall, reduction factors predicted from the model approach for early felling can reach two- to tenfold, whilst delayed felling can lead to reduction as high as 1.5–2-fold compared with the normal felling programme. However, changing standard forestry practices can lead to economic losses due to delays, or additional costs in harvesting the timber. Increased transport costs are also expected if nearby areas cannot be used as sources of wood for industry. Forestry operation plans may need to be approved by regional forestry administration. Markets may experience seasonal gluts and shortages so alternative wood sources should be available to compensate for the delay in harvesting. If large forest areas are clear-felled, the hydrology of the site must be considered especially with respect to possible erosion in the catchment (Nisbet et al. 2009).

5.3 Change of Hunting Season

Due to seasonal variation in diet of wild animals, the radiocaesium contamination of some game species varies significantly with season in Western and Eastern Europe. In particular, radiocaesium activity concentrations in muscle of game animals from areas where fungi can be abundant in certain years can be much higher than the average annual values (Hove et al. 1990; Johanson 1994). Hunting is normally restricted to certain periods of the year. By changing or restricting the hunting season to the time of year when the contamination levels in the game meat are not enhanced due to dietary preferences, the internal dose to humans consuming game meat can be reduced. A change of hunting season was applied in the FSU and some Nordic countries following the Chernobyl accident (IAEA 2006). Varying hunting times achieved a 2–3-fold reduction of radiocaesium activity concentrations in moose meat with even higher reduction of up to four to sixfolds for meat from roe deer, wild boar and wild reindeer. The effectiveness is greater if large quantities of mushrooms are available which occasionally happens in the autumn leading to potentially higher seasonal increases in wild meat contamination (Fesenko et al. 2012). The impact of changes in hunting season on breeding should be considered for each species. The continuous management of large game animals through hunting licenses is an integral part of controlling the populations of game animals at sustainable levels. It is thus important to continue hunting, or recommence hunting, as soon as possible after contamination occurs.

Some detrimental side effects may result from this measure. Increased grazing on adjacent agricultural lands may occur if the hunting season is delayed. If hunting is performed earlier than normal then lower slaughter weights may be anticipated. If hunting is restricted to seasons with bad weather, harsh climates may make hunting less attractive in some countries. The hunting season must not be changed to coincide with breeding and other seasons which are important for maintaining animal populations (Johanson 1994).

5.4 Optimisation of Forest Management

Logging may be transferred to areas with low external doses and to areas with lower radionuclide activity concentrations in wood. The same approach can be used for other forest products (Fesenko et al. 2012). Recommendations on suitable areas for tree felling and gathering of mushrooms or berries in forests of the FSU affected by the Chernobyl accident were given to local people and led to lower external doses to foresters or other people gathering food in the forest as well as substantial reductions in internal dose due to radiocaesium. The effectiveness depends on the contamination densities in forested areas. A reduction of the external dose of two- to threefolds may be achieved. For forest products (harvested wood, berries or mushrooms), reductions of two- to fivefold in internal doses are achievable (Fesenko et al. 2012).

Conclusions: Selecting Remedial Options in Areas Affected by Radiocaesium

The effectiveness of the available techniques used to reduce radionuclide activity concentrations in agricultural products varies depending on factors such as: application rates and duration of the application of remedial actions, the type of materials used, extent of adherence to required procedures (compliance), acceptability of the remedial options for the stakeholders and many other site specific factors to be taken into account within the remediation planning phase. Implementation of some remediation options may require specific equipment or resources which are not in normal usage in the production systems requiring remediation. Given the high variability in the effectiveness of remediation options, it is advisable to test their impact in relevant practical conditions in contaminated areas before using them on a wide scale (Howard et al. 2004, 2005; RIARAE 2006; Nisbet et al. 2009).

There are normally costs associated with implementation of remediation options which are readily defined for labour, consumables, equipment, transport and waste disposal. However, there are also other indirect costs of remediation such as the loss of production and retail sales through disruption and/or closure of businesses, loss of market share and regional impacts on tourism. Such indirect costs are often just as important as the direct costs, but more difficult to quantify partly because they can impact over much broader areas and populations than the contaminated area itself (Howard et al. 2005). Conversely, some remediation options can have direct monetary benefits such as additional crop yield due to mineral fertiliser application.

Some remediation options may produce contaminated by-products or so called "remediation waste". For example, if soil removal is implemented, a large quantity of contaminated soil may be generated even from relatively small areas. This may require long term disposal of the resulting waste and the cost of disposal may be much higher than that of implementing the remediation measure (Howard 2012). Waste disposal schemes for contaminated waste should preferably be planned before the waste is generated, with potential disposal sites and resources identified where possible (IAEA 2015). After the Fukushima Daiichi accident the removal of contaminated topsoil was widely used (IAEA 2015). There were many discussions regarding the planning and disposal of remediation waste to decide whether on-site or off-site disposal was preferable. As well as technical issues, aspects related to social considerations were important. There was a particular focus on perceptions of whether a "dilute and disperse" or "contain and concentrate" strategy was more justified (Oughton and Forsberg 2009).

To be a part of a remediation strategy the application of remedial options should be considered from many different aspects and be shown to be sustainable. This means that they need to sustain normal socio-economic activities and have no significant disadvantages such as unacceptable cost, waste generation or detrimental side effects. Some options require more rigorous analysis or additional experimental studies at selected sites and situations to that their applicability in a specific context can be defined before inclusion in a remediation strategy.

Phytoremediation represents an example of options which require additional consideration before application as a part of remediation strategy. The potential use of phytoremediation is based on the premise that some plants have a particularly high uptake of certain radionuclides. Therefore, if these plants are periodically harvested and disposed of in an appropriate manner, this option may provide the potential to remove radionuclides from the main environmental sink, the soil. However, currently there has been no small or large scale adoption of the methods at existing sites for radionuclides for three main reasons, namely: (1), the total amount of radionuclide removed from soil is a very small fraction of the total radionuclide content present, even for those radionuclides with a comparatively high transfer from soil to plant (2) the process would need to continue for many decades before the soil became adequately decontaminated to use for food production, and (3) the option generates waste which would then have to be disposed of appropriately generating additional costs.

Implementation of soil-based agricultural actions can change soil properties leading to direct environmental impacts including changes in biodiversity, soil fertility and structure and enhanced soil erosion. There can also be associated effects which reduce the quality of air and aquatic ecosystems. Conversely, some side effects of the application of agricultural remediation options can be beneficial. A good example is the increase in crop yield associated with enhanced fertilizer application. The increase in mass, apart from being a benefit to the farmer, may also dilute radionuclide activity concentrations in crops. Enhancing the productivity of grassland may also improve the condition of livestock and the effect may be particularly pronounced in previously less intensively managed systems.

Some *in-situ* biological and chemical leaching techniques could be applied to soil to remove radiocaesium such as soil washing, flotation, chemical extraction and bioleaching. Implementation of such options may require large quantities of the leaching substance, and can substantially contaminate surface and groundwater. Such options may have a substantial environmental detrimental effect, may prevent re-establishment of vegetation, and would not normally be a part of a sustainable remediation strategy for releases of radionuclides (IAEA 2015).

The imposition of restrictions on utilising forests can lead to negative side effects. If all access by the public is restricted, or limited, local residents may have insufficient access to resources such as wood collected for cooking, fires and other domestic and industrial purposes; berries and mushrooms; and access to forest meadows used for livestock. The cost of forestry may also be increased due to limitations on the time which forest workers can spend in contaminated forests which, in turn, can lead to deterioration in the maintenance of forests with negative ecological consequences. Application of restrictive remediation options such as limited access to forest in the areas affected by the Chernobyl accident had negative psychological and sociological consequences. The economic losses were also large (Salt and Rafferty 2001).

For many remediation scenarios, the application of remedial options can substantially change the ecological status of an area. The impacts, and hence acceptability, of deep-ploughing in an intensively agricultural area are high, but this will not also be the case for many semi natural ecosystems, even though the effectiveness may be much higher for the latter situation. In such cases, environmental legislation must also be taken into account (Nisbet et al. 2009).

Studies on effectiveness of remediation implementation in many areas affected by the nuclear accidents at the Chernobyl and Fukushima Daiichi accidents have identified many factors that had a highly detrimental impact on the economic and social activities on contaminated areas. Furthermore, some remediation actions may impose a stigma on the areas with contaminated environments (Oughton et al. 2004, 2009; IAEA 2015). The type of information disseminated about remediation should be targeted to meet a variety of needs of different stakeholders. The form of communication should be adapted to different levels of understanding and the prevailing circumstances to address the relevant issues, and should be implemented at the same time as the development of restoration strategies (IAEA 2015). Provision of local counting equipment may allow the public to monitor their own food, and improve autonomy and empowerment. The application of many remediation options can be limited by the wide range of factors discussed above which should be evaluated beforehand. Such evaluation needs to also consider administrative and regulatory issues such as legal constraints, environmental protection criteria; and cultural or heritage protection. In some cases there may be positive impacts in these areas. For instance, forest ecosystems may benefit from the absence of human exploitation (IAEA 2006).

Wide-scale implementation of remediation must take into account social and ethical issues when considering justification of possible options (Oughton et al. 2004; Oughton and Forsberg 2009). The consequences of implementation of remedial actions need to be evaluated with respect to potential impacts such as the effect on current and future generations, sustainability, and the relative harm to the environment compared with benefits to humans. If social aspects are respected, there is a greater likelihood that the implementation of remedial actions will be acceptable to both the public and operators (IAEA 2006; Fesenko et al. 2007; ICRP 2009). Thus, the impact on both individuals and communities needs to be considered. In particular, some remedial options may affect society by causing disruption (e.g. through restricting access or activities); anxiety and stress (e.g. by causing panic, upheaval); and stigma (e.g. by affecting businesses or tourism). Involvement of the affected communities in self-help options can reinforce liberty and dignity and be highly beneficial. These options were widely used in some regions of Belarus within the ETHOS project (Dubreuil et al. 1999). Remedial options implemented by the population themselves are normally ethically robust and often efficient and cost effective. Some of them do not require specific experience and can be carried out by local people with minimal training and advice (ICRP 2009). The involvement of affected population in implementation of remediation can give a feeling of control of the situation, which also prevents undue anxiety (ICRP 2009; IAEA 2015).

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