Thirty years after the Chernobyl accident: What lessons have we learnt?

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ABSTRACT

April 2016 sees the 30th anniversary of the accident at the Chernobyl nuclear power plant. As a consequence of the accident populations were relocated in Belarus, Russia and Ukraine and remedial measures were put in place to reduce the entry of contaminants (primarily 134–137Cs) into the human food chain in a number of countries throughout Europe. Remedial measures are still today in place in a number of countries, and areas of the former Soviet Union remain abandoned.

The Chernobyl accident led to a large resurgence in radioecological studies both to aid remediation and to be able to make future predictions on the post-accident situation, but, also in recognition that more knowledge was required to cope with future accidents. In this paper we discuss, what in the authors’ opinions, were the advances made in radioecology as a consequence of the Chernobyl accident.

The areas we identified as being significantly advanced following Chernobyl were: the importance of semi-natural ecosystems in human dose formation; the characterisation and environmental behaviour of ‘hot particles’; the development and application of countermeasures; the “fixation” and long term bioavailability of radiocaesium and; the effects of radiation on plants and animals.

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1. Introduction

At 01:23 on 26th April 1986 an experiment was started at number 4 reactor of the Chernobyl nuclear power plant in northern Ukraine (then part of the USSR). The purpose of the experiment was to investigate reactor safety in the event of failure of the mains electricity supply to the plant. Less than a minute after the start of the experiment there was a steam explosion which blew the lid of the reactor and resulted in the largest accidental release of radioactivity into the environment in the history of nuclear power production. The exposed reactor core continued to burn for approximately 10 days with continued releases of radioactivity into the atmosphere over this period.

As well as high contamination in the local area, radioactive plumes were transported over large areas of Europe with the highest depositions of radioactivity at distances from the NPP being due to wet deposition in rainfall.

Between the 27th April (when the about 44,000 inhabitants of Pripyat approximately 3 km from the NPP were evacuated) and 6th May the entire population of what has become known as the '30 km exclusion zone' around the NPP were evacuated. Initially a total of approximately 116,000 people were evacuated from an area of about 3500 km$^2$. Subsequently the number of evacuees rose to 350,000 within affected areas of Ukraine, Belarus and Russia. Many of these evacuated areas remain abandoned today.

A population of about 6 million people in Ukraine, Belarus and Russia were living in areas which were officially designated as 'contaminated' (>37 kBq 137Cs m$^{-2}$); 640 settlements with approximately 270,000 people had in excess of 555 kBq 137Cs m$^{-2}$ (Fesenko et al., 2006). Consequently, wide scale remedial actions were required in these former Soviet Union (SU) countries both in food production systems (Fesenko et al., 2007) and also to decontaminate some settlements (IAEA, 2006). Outside the SU, long-term remedial measures were put in place on animal production systems in a number of countries (Brynildsen et al., 1996; Åhman, 1999; Meredith et al., 1988; Wright et al., 2003). In Scandinavia the fallout necessitated a range of actions to protect the culture and lifestyle of the reindeer herding South Sámi people (Strand et al., 1989a; Stephens, 1987).

The Chernobyl accident led to a large resurgence in radioecological studies both to aid remediation and to be able to make future predictions on the post-accident situation, but, also in recognition that more knowledge was required to cope with future accidents. In this paper we discuss, what in the authors’ opinions, were the advances made in radioecology as a consequence of the Chernobyl accident.

This review paper is accompanied by an on-line virtual special issue in which the authors have selected 30 key papers published in Journal of Environmental Radioactivity which demonstrate the contributions of post-Chernobyl research (Beresford et al., 2016; http://dx.doi.org/10.1016/j.jenvrad.2016.01.023$^1$).

Descriptions of the Chernobyl accident and its consequences can be found in IAEA (2006) and Smith and Beresford (2005) (the latter being the source of information for the overview of the accident and aftermath given above).

2. Post-Chernobyl advances in radioecology

2.1. ‘Hot particles’

The release of highly radioactive fuel particles (generally referred to as ‘hot particles’) into the environment was a distinguishing feature of the radioactive contamination following the Chernobyl accident (Bogatov et al., 1990; Konoplev and Bobovnikova, 1990; Victorova and Garger, 1990; Sandalls et al., 1993; Salbu et al., 1994; Kashparov et al., 1996). The fuel particles were of either dense or loose structure and were composed of uranium oxides. The radionuclide composition of the fuel particles was similar to the fuel composition in the damaged unit with some depletion of volatile nuclides (131I, 134,137Cs, 106Ru, etc.). Sizes of deposited fuel particles ranged from hundreds of microns to a fraction of a micron (Sandalls et al., 1993). Within the 30-km exclusion zone, up to 10$^7$ particles m$^{-2}$ were observed (Victorova and Garger, 1990). Deposition of fuel particles decreased with increasing distance from the reactor site (Sandalls et al., 1993).

As a result of the breakdown of fuel elements and annealing of nuclear fuel, large amounts of volatile fission products (isotopes of I, Cs and others) were released into the atmosphere and partly condensed on inert particle carriers (Konoplev and Bobovnikova, 1990; Kashparov et al., 1996). These “condensation particles” had a lower activity concentration compared with fuel particles (Konoplev et al., 1993; Kashparov et al., 1996) and were similar to those found in global fallout after nuclear weapon tests and, therefore, their environmental behaviour could, generally, be predicted comparatively well. Nevertheless, behaviour of fuel particles within the environment was unstudied before the Chernobyl accident and, therefore, presented a serious scientific problem (Sandalls et al., 1993; Salbu, 2001).

Fallout from nuclear weapons testing had more than 90% of 90Sr and 137Cs in water soluble and readily exchangeable forms (Pavlotskaya, 1974; Konoplev and Bobovnikova, 1990). After the Chernobyl accident close to the NPP, the fraction of water soluble and exchangeable forms in the fallout was much lower, because of the presence of water-insoluble fuel particles, and depended on the distance from the damaged unit (Konoplev and Bulgakov, 1995). For example, the fraction of non-exchangeable 137Cs in the fallout near Chernobyl was about 75% (Konoplev and Bobovnikova, 1990; Bobovnikova et al., 1991), in the Bryansk region (Russia) it was 40–60% (Konoplev et al., 1996) and in Cumbria (UK) it was about 10% (Hilton et al., 1992). Because of the lower solubility of hot particles, the Chernobyl radionuclides had higher values of the distribution coefficient ($K_d$) in the “soil–water” system and, hence, slower migration and lower bioavailability in the area close to the NPP. Wash-off of dissolved 89Sr with surface run-off in the 30-km zone in 1986–1987 was lower than that after Kyshtym accident or nuclear weapon testing (Konoplev and Bobovnikova, 1990).

Mobility and bioavailability of radio.caesium in Western Europe was higher, and similar to global nuclear weapons testing fallout, because the particles deposited there were mostly condensation particles (Hilton et al., 1992; Konoplev and Bulgakov, 1995; Smith et al., 2000).

Apart from the differences in radionuclide speciation between Chernobyl and nuclear weapons testing fallout, the radionuclides also differed in their rates of change in availability in the soil. The mobility of radionuclides from nuclear weapons testing decreased with time because of fixation by soil components, while in the zone near the Chernobyl NPP, in the first years after the accident, the predominant process was leaching of radionuclides from fuel particles, which led to increased migration, especially for 89Sr (Konoplev et al., 1992; Kashparov et al., 1999, 2004; Konoplev and Bulgakov, 1999). In the first years after deposition, the uptake of 137Cs by plants in areas where aerosol fallout dominated was 4–5 times higher than that in areas with considerable hot particle contamination. However, subsequent transfer in areas with considerable hot particles exceeded that in areas dominated by aerosol fallout by three to five times (Fesenko et al., 1997). This effect has implications for the design and timing of remediation.

$^1$ The virtual special issue will be available on-line up to April 2017.
strategies.

A key advance of post-Chernobyl research was to use radionuclide speciation data to estimate the fraction incorporated in fuel particles (Konoplev and Bobovnikova, 1990; Konoplev et al., 1992; Kashparov et al., 1999, 2000, 2004; Konoplev and Bulgakov, 1999). The proportion of a radionuclide in fuel particles was assumed to be equal to the fraction of a radionuclide in non-exchangeable form minus the fraction fixed by the soil. In such calculations it is preferable to use data on 85Sr, which is more weakly fixed by soils than is 137Cs. Such data were used to obtain site and soil specific rate constants of radionuclide leaching from fuel particles \( k_l \); rates ranged from \( 10^{-3} \) to \( 10^{-4} \) day\(^{-1} \) (Konoplev and Bulgakov, 1999) or 0.05–0.3 year\(^{-1} \) (Kashparov et al., 2004) depending on location and soil type. Konoplev and Bulgakov (1999) observed dependence of \( k_l \) on the distance from the Chernobyl NPP (in the range 3–20 km) for the southern sector of the contaminated area — the leaching rate constant increased with distance. Based on statistical analysis of a large data set of dissolution rates, Kashparov et al. (2004) demonstrated dependence of \( k_l \) on the soil pH. Konoplev et al. (1992) used data on dynamics of radiocaesium speciation in soils of the 30-km zone to reveal the rate constant of its leaching and considered the rate of its subsequent fixation by clay minerals. The rate constants \( k_l \) for 137Cs and 85Sr were similar and indicated that disintegration of the particle controlled radionuclide release rates.

During the years immediately after the accident, fuel particles were primarily found in the upper few centimetres of soil both close to the reactor and at distances up to 250 km (Konoplev, 2001). Most of the particles were concentrated in the 0–1 cm layer and their proportion decreased markedly with depth. The vertical profile of radioactive particles in soil was found to be practically independent of the distance from the NPP and was mainly governed by the soil type. The lack of dependence of the vertical distribution of particles on their size and chemical nature results from the fact that the primary mechanism of long-term radionuclide migration in the upper soil layer was mixing by soil flora and fauna.

In soils, fuel particles had virtually disintegrated within 10 years (Konoplev and Bulgakov, 1999). The opposite situation was observed in the cooling pond of the Chernobyl NPP, where the vast majority of long-lived radioactivity was deposited as fuel particles. In the cooling pond sediments, the majority of 85Sr activity still occurs in the form of fuel particles (Bulgakov et al., 2009). Due to low dissolved oxygen concentration and high pH, dissolution of fuel particles in the cooling pond sediments is significantly slower than in soils. However, at the present time these conditions are changing since the pond is being drained and a significant part of the sediments are being exposed to the air. This significantly enhances the dissolution rate of fuel particles in exposed sediments, and hence, it is anticipated that the mobility and bioavailability of radionuclides will increase with time (Bulgakov et al., 2009). Model calculations have shown that in newly exposed sediments fuel particles will be almost completely dissolved in 15–25 years, while in parts of the pond which remain flooded, fuel particle dissolution will take about 100 years (Bulgakov et al., 2009).

2.2. Behaviour in soils and sediments

The mobility and bioavailability of radionuclides of accidental origin in the terrestrial and aquatic environment are governed by their chemical forms in fallout and site-specific environmental characteristics which determine rates of leaching, fixation-remobilisation, as well as sorption-desorption of the mobile fraction (its solid-liquid distribution).

After deposition on the surface of ground and water bodies, radionuclides are subject to physicochemical and biological processes which leads to changes in their speciation. Dissolved radionuclide is adsorbed on the solid phase by ion exchange. Exchangeable radiocaesium transforms into the non-exchangeable (fixed) form (Comans et al., 1991; Hird et al., 1995; Konoplev and Bulgakov, 1995, 2000). Fuel particles become oxidized and disintegrates, and radionuclides incorporated in fuel particles transfer to solution.

It is now well established that the high retention of radiocaesium in soil and bottom sediments is largely determined by two different processes: fixation and reversible selective sorption (Sawhney, 1972; Cremers et al., 1988; Hird et al., 1995; Konoplev et al., 2002). Fixation describes the “permanent” (or at least long-term) replacement of interlattice K- by Cs-ions due to collapse of expanded edges of mineral's crystal lattice interlayers, Hird et al. (1995) found that soil organic matter can stabilise interlayers in their expanded state and reduce Cs-fixing ability of soil even though its Cs-fixing potential is high.

The equilibrium between dissolved and exchangeable forms is achieved relatively rapidly and in many cases it is reasonable to consider them as a single mobile form (Konoplev et al., 1996). Fixation of radionuclides is the transfer from their mobile form to fixed form. Models were proposed in which radiocaesium fixation is treated as an irreversible process (Comans and Hockley, 1992). The data available suggest that at the initial stage of fixation and for processes of a relatively short time-scale, this assumption is warranted. However, the data for long-term transformation of chemical forms of radionuclides in soil after nuclear weapon testing (Pavlotskaya, 1974), the Kyshtym accident (Konoplev and Bobovnikova, 1990) and in the long-term after the Chernobyl accident indicate the existence of a remobilization process that is the reverse of fixation (Konoplev et al., 1992; Smith and Comans, 1996; Konoplev and Bulgakov, 2000; Smith et al., 2000b). After deposition of radiocaesium onto the soil, the fraction of its exchangeable form does not decrease to zero, as should in theory happen during irreversible fixation, but it decreases to a certain level, independent of the amount of radionuclide applied, and then does not change.

\[ K_d = \frac{\text{activity in soil}}{\text{activity in solution}} \]

Fig. 1. Conceptual model of radiocaesium bioavailability in soil/soil-plant transfer; \( K_d \) is defined as the ratio of the concentration of radionuclide reversibly sorbed by the solid phase to its concentration in the liquid phase.
significantly because of an equilibrium steady state between fixation and remobilization (de Koning and Comans, 2004; Konoplev and Bobovnikova, 1990; Konoplev and Bulgakov, 1995; Konoplev et al., 1996).

Reversible selective sorption of radiocaesium occurs on frayed edge sites (FES), located at the edges of micaeous clay particles (Cremers et al., 1988). The ability of a solid to sorb radiocaesium selectively is characterised by the capacity of the selective sorption sites (FES) or by the so-called radiocaesium interception potential (RIP), which is the product of the FES capacity and the selectivity coefficient of radiocaesium in relation to the corresponding competitive ion (Sweeck et al., 1990). Cremers and co-workers (Cremers et al., 1988; Sweeck et al., 1990) developed a method for the quantitative determination of FES capacity and RIP. The method is based on using silver thiourea (Ag(TU)\textsuperscript{+}) as a masking agent for regular exchange sites (RES) which correspond to the planar and easily exchangeable sites. RES selectively bind Ag(TU)\textsuperscript{+}. At the same time Ag(TU)\textsuperscript{+} does not interact with FES, because of the molecule’s large size, and the approach allows the study of caesium sorption—desorption on FES.

The $^{137}$Cs concentration in the soil solution is the key characteristic determining its uptake by plants (Konoplev et al., 1993). Under the assumption that radiocaesium uptake by plants is proportional to the fraction of radionuclide in the root exchangeable complex, which is dependent on the composition of soil solution (Smolders et al., 1997), models of radiocaesium bioavailability in soils have been proposed (Konoplev et al., 1999, 2000; Absalom et al., 2001). This has allowed the parameterisation of radiocaesium soil-plant transfer through soil characteristics (Fig. 1).

2.3. Exposure pathways from food consumption

In the first few weeks after the accident, $^{131}$I was the main contributor of dose to the population in Belarus, Russia and Ukraine. An initial lack of advice to private producers (i.e. not the state-owned collective farms, but owners of cows largely for self-consumption) of milk on how to reduce the $^{131}$I contamination of state-owned collective farms, but owners of cows largely for self-consumption. Ukraine. An initial lack of advice to private producers (i.e. not the population subsystems, the contamination of forest and game from semi-natural ecosystems (upland pastures, forests) (Desmet and Myttenaere, 1988) as are some berries (Beresford et al., 2001; Calmon et al., 2009). Whilst initially after the Chernobyl accident the contribution of agricultural products (mainly milk) dominated the internal dose, in the longer-term the importance of fungi (wild berries in the decomposition of the population subsystems) increased. This was partially because of the long-ecological half-lives of radiocaesium in berries and fungi and the implementation of countermeasures in agricultural systems (and hence a reduction of doses received via this pathway), but, there are also reports of an increased consumption of berries and fungi with time after the accident (Fensenko et al., 2001; Travnikova et al., 2001). For a Russian village Travnikova et al. (2004) estimated that the contribution of fungi and berries to the total dietary radiocaesium intake increased from 6% in 1987 to 25% in 1996. Mean intakes of forest berries and wild fungi of 2.4 and 7.2 kg a\textsuperscript{-1} were recorded for four settlements in Russia (Mehli, 1998).

Having surveyed inhabitants of six rural settlements in Belarus, Russia and Ukraine, Strand et al. (1996) reported that: 40–75% of interviewees consumed wild fungi, forest berries were consumed by 60–70% and; fish from local lakes by 20–40%. For a lake side village in Russia, Travnikova et al. (2004) report that 50–80% of the $^{137}$Cs intake came from fungi and fish, with 30–50% of the total $^{137}$Cs intake coming from lake fish. Freshwater fish was also found to contribute 20–40% of the radiocaesium intake by rural populations in Norway (Strand et al., 1989b).

Within rural populations in Russia, Skuterud et al. (1997) observed a 60–70% increase in whole body radiocaesium activity as a consequence of fungi consumption in autumn. Correlations were also reported between the consumption of fish and forest berries and $^{137}$Cs whole body measurements in Russian settlements (Strand et al., 1996), though, there were also correlations between the intakes of forest berries, freshwater fish and fungi. Shutov et al. (1996) estimated that fungi and berries could contribute up to 60–70% of dietary $^{137}$Cs intake of adults. For one Ukrainian village Beresford et al. (2001) present data showing milk and fungi contributed approximately 60% of the daily $^{137}$Cs intake in June/July when fungi (and forest berry) intake would not be expected to be high (fungi and forest berries being most prevalent in autumn). Outside of the FSU the focus has been on high radiocaesium concentrations in livestock and game from semi-natural ecosystems (upland pastures, forests) (Desmet and Myttenaere, 1988) where a high $^{137}$Cs soil-plant transfer was persistent in organic soil types (Hird et al., 1995; Rigol et al., 2002). Long ecological half-lives in excess of ten years and in some instances tending towards the physical half-life of $^{137}$Cs in the longer-term were observed in a number of species (e.g. reindeer (Skuterud et al., 2005), roe deer (Zibold et al., 2001) wild boar (Pröhl et al., 2006)), Seasonal peaks in
late summer/autumn have been observed in a number of species as a consequence of consuming relatively highly contaminated mushrooms (e.g. Hove et al., 1990; Zibold et al., 2001). The situation for reindeer herding South Sámis in central Sweden and Norway deserves special mention. Depending upon natural pastures for their reindeer herds, and largely on reindeer meat, freshwater fish and wild berries in their diet, the Chernobyl fallout suddenly threatened their whole life-style (Stephens, 1987). A suite of actions was implemented to maintain their lifestyle, including dietary advice, economic support for purchase of alternative foodstuffs, and whole-body monitoring as a control of doses (Strand et al., 1992b; Mehli et al., 2000). These actions reduced ingestion doses during the first years by nearly 90%, however, consumption of reindeer meat still comprised about 90% of their radiocaesium intake (Strand et al., 1992b; Skuterud and Thørring, 2012). Since the mid-1990s onwards the whole-body radiocaesium levels in the South Sámis of central Norway have been comparable to those of rural populations close to Chernobyl (Mehli et al., 2000; Skuterud and Thørring, 2012).

Given that transfer through animal products was a major route of exposure, there was a considerable effort devoted to understanding the metabolism of radionuclides by farm animals (see, for instance, special issue of Journal of Environmental Radioactivity on this topic (Beresford et al., 2007)). An understanding of the metabolism in farm animals can better inform on the practicality and implementation of remedial measures (e.g. Ahman, 1999; Voigt and Kiefer, 2007; Howard et al., 1987). Key advances made in animal radioecology included the use of faecal marker approaches to estimate herbage (and hence radionuclide) intake in grazing animals (Mayes et al., 1994; Beresford et al. 1989) and the application of dual isotope techniques to determine absorption of radionuclides from the diet (Mayes et al., 1996; Cooke et al., 1996), including under field conditions in the Chernobyl exclusion zone (Beresford et al., 2000).

2.4. Remediation

Deposition of radionuclides from the Chernobyl led to the long-term implementation of remedial measures in the FSU and other European countries. Here we consider those methods investigated, developed and used to remediate affected areas (see also special issue of Journal of Environmental Radioactivity (Voigt, 2001)).

2.4.1. Remediation in Ukraine, Belarus and Russia

In the most seriously contaminated areas of the FSU, the consumption of foodstuffs containing $^{137}$Cs (and initially $^{134}$Cs was the major source of the public exposure (Jacob et al., 2001; IAEA, 2006) with milk and other animal products representing a major exposure pathway to humans (Fesenko et al., 2007).

Because the accident occurred in the latter part of spring, there was little scope to remove animals from pasture and feed uncontaminated feed (as silage/hay stocks are at their lowest in spring). Maximum radionuclide activity concentrations in agricultural products occurred in the most affected areas of the FSU in the first days to weeks after the accident, reaching hundreds of thousands of Bq per kilogram or litre (IAEA, 2006). In the first weeks the main remedial objective was to prevent consumption of contaminated milk and to decrease milk $^{137}$Cs activity concentrations (Fesenko et al., 2007). Measures implemented were: (i) radiation monitoring at processing plants and rejection of milk with $^{137}$Cs activity concentrations above action levels (3700 Bq l$^{-1}$ at that time); (ii) processing rejected milk (mainly converting milk to storable products such as condensed or dried milk, cheese, or butter); (iii) indoor feeding of uncontaminated feed (though as already noted feed availability was limited). Processing milk avoided considerable economic losses (Alexakhin et al., 1996). In the most contaminated areas, a ban was imposed on keeping livestock. Delayed harvesting of forage and food crops was used as a method of reducing dietary radionuclide intake. During the first period the countermeasures were largely directed towards "collective" milk.

By the end of 1986, only four regions of Russia (Bryansk, Tula, Kaluga, and Oryol), five regions of Ukraine (Kiev, Zhitomir, Rivno, Volyn and Chernigov) and three regions of Belarus (Gomel, Mogilyov and Brest) were producing food (largely animal) products exceeding action levels for radiocaesium. Therefore, from 1987, application of countermeasures to reduce the $^{137}$Cs activity concentrations in milk and meat was the key focus of remediation strategies (Alexakhin, 2009).

The priority was amelioration of soil with the aim to reduce radionuclide transfer to plants, with special attention paid to those areas used for production of animal fodder. Soil amelioration options included: ploughing, liming, application of mineral fertilisers and, 'radical' or 'surface' improvement of meadows.

Ploughing was only feasible in areas with fertile soils with a deep organic horizon, both deep and shallow ploughing was used extensively. The effectiveness of ploughing was a long-term reduction in radiocaesium transfer to plants by a factor of 2–4 (Fesenko et al., 2007), a reduction of 20 fold could be achieved using skim and burial ploughing. Based on research carried out in the first years after the accident the application rate of lime for contaminated regions was increased, over that normally used in agricultural production, by approximately 1.5 times depending on soil pH, soil type and crop. The amount applied every four-five years typically ranged from 2 to 10 t ha$^{-1}$ resulting in reductions in the $^{137}$Cs and $^{90}$Sr transfer to crops by a factor of 1.5–3.0 (Fesenko et al., 2007). The use of nitrogen, phosphorus and potassium fertilisers reduces the Cs:K ratio in the soil solution and also provides ideal crop growth conditions. The optimum N:P:K ratio to achieve the maximum reduction (by a factor of 3 (Fesenko et al., 2007)) in root uptake of radiocaesium was found to be 1:1.5:2 (RIARAE, 1991).

'Radical' improvement was a key measure carried out extensively in practically all contaminated areas of Belarus, Russia and Ukraine. This option included removing vegetation, ploughing, liming, fertilization and reseeding and has been effective in field conditions achieving a reduction in root uptake of radiocaesium of 2–3 fold. For organic soils the effectiveness increased to 3–5 fold, with maximal effectiveness of 10–15 fold for wet peat soils when drainage was also conducted. The mechanism of action and efficiency of radical improvement of forage-land and pastures depended on the types of meadow and soil properties (Sanzharova et al., 1996; Vidal et al., 2001). 'Surface' improvement was applied to sites with natural vegetation and low productivity (e.g. floodplain meadows) and included reseeding with different grass species with the land being more regularly managed (Sanzharova et al., 1996). With time, repeated management of previously treated soils was necessary. Whilst the most appropriate application rates could be defined, in reality, rates of application were sometimes financially constrained.

Options such as changing arable land into meadow and 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: oil for human consumption and protein cake as an animal fodder (Bogdevitch et al., 2002). The varieties of rape grown were selected for their low uptakes of caesium and strontium.

One of the most important and frequently used countermeasures for meat production systems was 'clean feeding', the provision of uncontaminated feed for a period prior to slaughter (Bogdevitch et al., 2002).
Use of caesium binders to reduce absorption of caesium from the gastrointestinal tract of livestock in the FSU did not become a widespread action until 1994, as a result of international co-operation sharing the positive experiences with the use of such binders in western Europe (in particular Norway, see later) (IAEA, 1997). The most effective Cs binders are hexacyanoferrate compounds often referred as ‘Prussian Blue’. Prussian Blue application has been especially effective in settlements where there was a lack of meadows suitable for radical improvement. In Belarus, a special concentrate with Prussian Blue was produced and distributed at a rate of 0.5 kg of concentrate per cow daily and an average reduction factor of 3 for milk was achieved. However, the use of imported hexacyanoferrates was prohibited as a consequence of their high cost. A local produced hexacyanoferrate, called ferrocyn was used instead (Howard et al., 2001). Delivery mechanisms included: pure powdered compound; rumen dwelling boli (see below); saltlicks; and as bifege, sawdust with 10% absorbed ferrocyn (Ratnikov et al., 1998).

The maximum effect from countermeasures application was achieved in 1986—1992 (Fesenko et al., 1997). Since 1991, the proportion of animal products with radocaesium activity concentrations exceeding action levels has been <10% of the gross output from contaminated areas. Because of this, the use of agricultural countermeasures was drastically reduced and their application rates were inadequate not only as a countermeasure but also for conventional food production; some increase in 137Cs transfer was observed as a consequence (Fesenko et al., 2001).

The rates of decrease of 137Cs concentrations in foodstuffs, especially in milk, differed significantly across the affected regions and over time after the accident. This was primarily because soil-based countermeasures as radical improvement could not be implemented for the entire contaminated area as this would have led to a shortage of pasture. Therefore, the effectiveness of agricultural countermeasures increased from 1987 to 1992 when practically all affected areas were optimally remediated.

The extent to which the different remediation measures were used varied between the three countries. Over the first 20 years after the accident the implementation of agricultural countermeasures averted 30—40% of the internal collective dose (excluding thyroid dose) or around 20—25% of total collective dose that would be received by the residents of contaminated areas of Russia, Ukraine and Belarus without the use of countermeasures (Fesenko et al., 2007). Analyses of possible strategies for remediation of rural areas affected by the Chernobyl accident showed that even two decades after the Chernobyl accident application of remedial actions was still cost-effective. Thus, further remediation of affected areas would result in considerable reduction in the exposure of the population (Fesenko et al., 2007).

As discussed above, since the Chernobyl accident, it has been widely recognised that natural products such as forest fungi and berries, freshwater fish and privately produced milk can substantially contribute to the radiation dose received by humans especially to rural populations within the FSU. Whilst consumption of such foodstuffs was banned in some areas, these bans were often ignored (IAEA, 2006). It was therefore recognised that affected populations could be given guidance on how to reduce their radocaesium intake (such guidance aimed at groups such as hunters, fishermen and the Sámi reindeer breeders was effectively implemented in 1986 in Norway (Strand et al., 1992b)). Beresford et al. (2001) suggest advice on how the local population could themselves reduce their radiological risk from the consumption of fungi, berries and private milk and how this could be presented in an understandable manner. Lepicard and Hériard Dubreuil (2001) and Lochard (2007) report on work in a Belarusian settlement to build a practical radiological know how within the community such that the villagers could begin to manage and improve their own radiological situation. In Lepicard and Hériard Dubreuil (2001), evidence is presented on how this reduced contamination in milk and also gave the villagers economic benefit as the reduced contamination also allowed them to sell their milk to the regional dairy.

The application of countermeasure against external exposure was also of importance in many Russian and Belorussian settlements (Jacobsen et al., 2001). The paper by Roed and Andersson (1996) provides results of research on the application of a variety of decontamination techniques in the urban environments. It was shown that even several years after the accident, when the work was conducted, the effectiveness of most of decontamination options was still high and up to 75% of 137Cs could be removed at relatively little cost.

In the affected FSU countries, forests are not only an economic resource, but also a source of many social and cultural activities which can be also affected by the contamination. A number of factors have to be considered when assessing the applicability of forest countermeasures: radiological, economic, environmental/ecological and social. For instance, economic and social consequences may be incurred in addition to the expense of countermeasure implementation. The paper by Shaw et al. (2001) provides an example of a cost—benefit analysis applied to determine the cost effectiveness of several management strategies potentially applicable to the contaminated forests. The management strategies were considered as single options and in likely combinations. Only the banning of fungi collection and restriction of public access proved to be cost-effective management and only at relatively high levels of 137Cs contamination. No potential countermeasure was assessed to result in a cost-effective saving of doses to forest workers.

2.4.2. Extensive animal production systems

Outside of the FSU there were long-term problems of animal derived food products having 137Cs activity concentrations exceeding permissible levels for entry into the human foodchain in Scandinavia and the UK. The animals affected in these ecosystems are often free-ranging and many are infrequently handled (e.g. sheep, semi-domesticated reindeer). Long-term radocaesium contamination of game animals was also observed in a number of countries (e.g. Johanson and Bergström, 1994; Semizhon et al., 2009; Strebl and Tataruch, 2007; Vilic et al., 2005; Zibold et al., 2001).

Reindeer husbandry in central Sweden and Norway was particularly affected by the Chernobyl fallout, and both countries chose to increase their permissible levels for radocaesium in traded reindeer meat to 1500 Bq kg−1 for 137Cs and 6000 Bq kg−1 for 134:137Cs, respectively. The aim of this was to reduce the consequences for the South Sámi reindeer herders (Ahman, 1999; Strand et al., 1989). In addition, a change in slaughter season was introduced to avoid the highest contamination levels.

Prior to the Chernobyl accident a number of feed additives had been identified which if administered to farm animals would effectively reduce absorption from the gastrointestinal tract (GIT); typically about 80% of ingested Cs is absorbed by ruminants (Howard et al., 2009). These additives included bentonite (van den Hoek, 1976), vermiculite (Hazzard, 1969) and hexacyanoferrate compounds (Giese, 1989). Consideration of the use of these...
compounds prior to Chernobyl had relied upon access to the affected animals such that the binder could be incorporated into their daily feed. Such an approach was used as a remedial measure following the Chernobyl, for instance, in Sweden and Norway where the clay mineral, bentonite, was added to the feed of corralled reindeer and dairy animals (Hove, 1993; Ahman, 1999; Howard et al., 2001). Hove (1993) reported a 50% reduction in radioocaesium transfer to milk and meat with bentonite at an administration rate of about 500 mg bentonite kg\(^{-1}\) body mass (BM), with a maximum reduction of 80% at an intake rate of 2 g bentonite kg\(^{-1}\) BM. Bentonite was added to the diet of dairy cattle and goats in contaminated areas of Norway at a rate of 5–10% of the feed by mass as a post-Chernobyl remediation measure (Hove, 1993).

However, whilst it has the advantage of being relatively inexpensive, bentonite was not suitable for all circumstances. It could not be administered to the free-ranging animals which were a challenging long-term issue in areas affected by the Chernobyl accident outside of the ISU. In the UK, the repeated application of bentonite to pastures was investigated as a delivery mechanism and this decreased the radioocaesium activity concentration in sheep meat. However, the reduction in radioocaesium transfer was accompanied by a decrease in the intake of herbage (measured using faecal marker techniques) by adult sheep with a concomitant loss of body weight; the approach was also impractical for application over the large areas of rugged terrain affected (Beresford et al., 1989). Bentonite administration rates above 500 mg bentonite kg\(^{-1}\) BM (about 2% of the feed) to reindeer reduce appetite when fed as powder (Hove et al., 1991), and was not recommended because of increased water requirements (which could be challenging during winter) and the unspecific binding of essential elements and electrolytes (Ahman et al., 1990; Ahman, 1996).

Hexacyanoferrates are much more efficient Cs binders than clay minerals. After the Chernobyl accident NH\(_4\)-iron-hexacyanoferrate (AFCF, Giese salt) was authorised for administration to farm animals initially in a small number of countries (Norway, Sweden, Germany and Austria) (Hove, 1993) and later across the European Union (Howard et al., 2001). In studies investigating post-Chernobyl remediation measures, reductions in the radioocaesium transfer to reindeer of 60% were observed at administration rates of 15% AFCF, 75% BaSO\(_4\) and 10% beeswax (Hove and Solheim Hansen, 1993). The high density of the boli means that they are retained in the reticulorumen where abrasion causes a gradual release of AFCF; three boli were administered per animal. The boli were subsequently modified by the addition of a wax coating to delay the start of bolus degradation and hence prolong their active life resulting in a greater reduction in the radioocaesium activity concentrations of animals at the time of slaughter (Solheim Hansen et al., 1996). In Norway the use of AFCF-boli was considered a cost effective remediation strategy (Brynildsen et al., 1996). The application of the Norwegian boli was considered in the UK. However the bolus was found to be too big to be administered to lambs of the native, small upland breeds. Smaller boli containing higher AFCF concentrations were manufactured and tested (Beresford et al., 1999). The maximum concentration of AFCF was found to be 20%, concentrations higher than this resulted in too rapid bolus degradation. Smaller boli (14 mm diameter by 50 mm length) containing 20% AFCF were subsequently tested on a hill farm with a reduction of >30% in the radioocaesium activity concentration in meat being observed 51 d after the administration of 3 boli per animal (Howard et al., 2001).

The other AFCF delivery method developed and used for free-ranging animals was the incorporation of AFCF (2.5%) into salt licks. In Norway it was estimated that a 50% reduction in the radioocaesium activity concentration of animals was achieved (Hove, 1993). However, effectiveness is dependent upon how frequently a given animal uses a salt lick and hence in a given flock/ herd effectiveness will be variable. Salt-licks would not be an effective remediation measure in coast areas where animals have no need for supplementary Na.

Other successful remediation measures employed after the Chernobyl accident included the altering of slaughter times, feeding uncontaminated feed prior to slaughter and the use of live-monitoring, and the controlled entry of animals into the foodchain (Brynildsen et al., 1996; Howard, 1993; Ahman, 1999).

### 2.4.3. Freshwaters

Following a radioactive fallout, there are a number of possible intervention measures to reduce radioactive doses to the public via the surface water pathway (Fig. 2) (Smith et al., 2001; Monte et al., 2009). After the Chernobyl accident, it was shown (Voitsekhovitch et al., 1997) that doses from terrestrial foodstuffs are in general more significant than doses from drinking water and aquatic foodstuffs. Aquatic countermeasures were, however, used and

![Fig. 2. Freshwater dose pathway indicating potential intervention measures. Dashed lines indicate pathways of lower potential importance (Smith et al., 2001).](image-url)
tested in response to high activity concentrations in surface waters during the first few weeks and subsequently high long-term contamination of freshwater fish; they were also used as a method of public reassurance.

The most effective and viable measures to reduce radioactivity in drinking water were found to be those which operate at the water treatment and distribution stage (Smith et al., 2001). Temporary bans on drinking water were put in place and switching or blending of supplies with less contaminated waters was used effectively (Tsarik, 1993; Voitsekhovitch et al., 1997; Voitsekhovitch, 1998). In the Dnieper Waterworks Station, activated charcoal and zeolite were added to water filtration systems to remove, respectively, 131I and 106Ru, and 137Cs and 90Sr (Tsarik, 1993).

Intervention measures to reduce concentrations of radioactivity in rivers and reservoirs were much less efficient at subsequently reducing doses via the drinking water pathway. Dredging of canal-bed traps to intercept suspended particles in rivers (Voitsekhovitch et al., 1988) and zeolite containing dykes were found to be very ineffective: only 5–10% of the 90Sr and 137Cs in the small rivers and streams were adsorbed (Voitsekhovitch et al., 1997; Voitsekhovitch, 1998). The construction of a dyke around the most contaminated areas of the Pripyat flood plain proved effective in reducing 90Sr loads during flood events (Voitsekhovitch et al., 1997).

Bans on consumption of freshwater fish were applied in some lakes in the FSU (Ryabov, 1992), though these were not always adhered to. Similar bans were also put in place in Scandinavia (Brittain et al., 1991; Tveten et al., 1998). Lake timing to reduce activity concentrations in fish were found to be ineffective for radio caesium (Håkanson and Andersson, 1992), though addition of potassium to lakewaters appears promising in some situations (Smith et al., 2003).

2.5. Effects on wildlife

The impact of very intense radiation in areas of forest to the west of the reactor resulted in dramatic early ecosystem impacts of the accident (Arkhipov et al., 1994; Kryshev et al., 2005). Coniferous trees were killed in an area of forest covering 4 km2 (approximately 100 000 m2) and were also used as a method of public reassurance. The most effective and viable measures to reduce radioactivity in drinking water were found to be those which operate at the water treatment and distribution stage (Smith et al., 2001). Temporary bans on drinking water were put in place and switching or blending of supplies with less contaminated waters was used effectively (Tsarik, 1993; Voitsekhovitch et al., 1997; Voitsekhovitch, 1998). In the Dnieper Waterworks Station, activated charcoal and zeolite were added to water filtration systems to remove, respectively, 131I and 106Ru, and 137Cs and 90Sr (Tsarik, 1993).

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Early radiation damage to other plants and animals is less well documented, though declines in populations of small mammals and reduced embryo survival were observed in the early period (Geras’kin et al., 2008; Hinton et al., 2007). Many domestic animals were evacuated from the Exclusion Zone, but radiation damage was observed in some which remained. Thyroid doses from 131I of up to 200 Gy resulted in death of some horses and cows due to radiation damage to the thyroid tissue (Shigematsu et al., 1991).

Studies of pine trees (Arkhipov et al., 1994) reported that by 1991 pine (Pinus sylvestris) trees had returned to “normal” with no obvious morphological damage in stands in the low and medium (up to 10 Gy) dose regions, and that a new plant community was developing in the severely damaged regions. However, in 1995 there was still evidence (Geras’kin et al., 2003) of cytogenetic damage in seedling roots of pines in two sites at Chernobyl of dose rates (approximately 0.25 and 27 µGy h−1). However, it is not clear whether this was a result of the initial acute dose (10–20 Gy at the high dose site) or the ongoing chronic exposure, or a combination of the two. A reduction in mutation load with time (Abramov et al., 1995) and adaptation (Kovalchuk et al., 2004) was observed in studies of Arabidopsis thaliana. Abramov and coworkers (Abramov et al., 1995), found that mutation load was increased in the first two years after the accident but that “in areas with contamination up to 10 mR h−1 (approximately 100 µGy h−1) the mutation load decreased to control levels by 1990. In the areas with contamination up to 130 mR h−1 (approximately 1300 µGy h−1) the mutation load (still) exceeded the control by 4–8 times”.

Initial effects on soil dwelling invertebrates were severe in the most contaminated areas (Krivolutsky et al., 1999; Krivolutskii and Pokarzhievskii, 1992): doses of about 30 Gy resulted in “mortality of eggs and early life stages” of invertebrates (Hinton et al., 2007). This led to a major decline in biodiversity in the most contaminated forest soils; this did not recover until 1995 (Krivolutsky et al., 1999).

In the longer term, a study (Møller and Mousseau, 2009) observed dramatic declines in abundance of dragonflies, bees and butterflies at dose rates as low as 1 µGy h−1, two orders of magnitude lower than the level at which significant effects on populations of invertebrates would be expected to be found (UNSCEAR, 1996) and in the range typical of natural background exposures (Beresford et al., 2008). Recently, Lecomte-Pradines and co-workers (Lecomte-Pradines et al., 2014) observed little effect of radiation on soil nematode assemblages at Chernobyl at dose rates up to 200 µGy h−1. An observed change in community structure at high dose rate sites was attributed either to ongoing chronic radiation, or to the long term effects of the initial impacts of the accident on invertebrate communities.

Prior to the Chernobyl accident, mammals were perceived to be among the most radio-sensitive taxonomic group. A series of studies of rodents in highly contaminated sites (average dose up to 830 µGy h−1) found no clear evidence of genetic change compared to control sites (Baker et al., 2001, 1996b, 1997; Wickliffe et al., 2002), although a study in Belarus at much lower dose rates (up to 30 µGy d−1) found an increased frequency of chromosome aberrations (Goncharova and Ryabokon, 1995). At the population level, Baker and co-workers (Baker et al., 1996a) found no difference in the diversity and abundance of small mammal populations in the most highly contaminated areas compared to control sites outside the exclusion zone. Studies of larger mammals using snow track surveys (Møller and Mousseau, 2013) reported significant population declines in mammal abundance with increasing contamination. A study covering a much larger area (Deryabina et al., 2015), however, found no relation between radioactive contamination density and mammal abundances.

Though there was no evidence of mortality of aquatic species after the accident, significant damage to the reproductive systems of fish was observed. Increased sterility and gonad abnormalities of silver carp in the Chernobyl Cooling Reservoir was observed from 1989 to 92 at cumulative dose rates of order 10 Gy (Belova et al., 1993; Makeeva et al., 1994). Increased aberrations in reproductive cells of fish were still observed in the period 1999–2004 in the NPP cooling pond and the highly contaminated Glubokoye Lake (Belova et al., 2007). An increased rate of DNA strand breaks was observed in fish from the cooling pond compared to a control site (Sugg et al., 1996). A study of pond snails (Lymnaea stagnalis) in the contaminated Perstok Lake in Belarus in 2004 (Golubev et al., 2005) observed an increase in micronuclei in the cells of contaminated snails, though the embryonic mortality in the contaminated lake was lower than at a control site. So far there have been no population studies of fish in lakes around Chernobyl, largely owing to the difficulty of such studies. A study of invertebrates (Murphy et al., 2011), however, observed no decline in abundance or diversity in invertebrate communities in contaminated lakes at dose rates up to about 30 µGy h−1.

There is little information on the effects of radiation on birds in
the early years after the accident, but there have been a number of studies of long term effects. Observed radiation effects have included increased morphological abnormalities (Møller et al., 2007); increased proportions of non-breeding birds at contaminated (ca. 3–50 μGy h⁻¹) sites (Møller et al., 2005a); elevated frequency of cataracts (Mousseau and Møller, 2013); and reduced abundance (Møller and Mousseau, 2007). The damage mechanism has been attributed by these authors to increased oxidative stress caused by production of free radicals by radiation interaction within cells (Møller et al., 2005b), though this mechanism has been questioned at the dose rates currently pertaining in the Chernobyl Exclusion Zone (Smith et al., 2012).

The early severe impacts of radiation in the most contaminated parts of the Exclusion Zone (at extremely high dose rates) are clear. But 30 years on, the radioecology community has yet to reach a consensus on the effects of longer term chronic low dose radiation on biota at Chernobyl. Dose rates to animals and plants in the most contaminated areas are still above 40 μGy h⁻¹ (1 mGy d⁻¹), within the range of dose rates where some radiation damage would have been expected (UNSCEAR, 1986). It is important to note that these high dose rates are found in only a small fraction (around 1–5%, depending on species) of the Exclusion Zone surface area. Nevertheless, studies, particularly on birds, have apparently observed severe effects at much lower dose rates.

It has been hypothesised (Garner-Laplace et al., 2013; Møller and Mousseau, 2011) that biota living in natural conditions are more susceptible to radiation than in laboratory conditions, leading to effects at lower levels than would be expected from laboratory experiments. This is a plausible hypothesis, yet there remains significant controversy over the strength of empirical evidence to support it. It is as yet not clear which observed effects are directly linked to radiation, what dose rates they are observed at, the mechanism for radiation damage and what the ecological significance of this damage is. This controversy is in large part due to the complexity of the problem — there are many different wildlife species exposed to different (and often uncertain) radiation dose rates, and there are many different potential effect endpoints, ranging from markers of DNA damage up to animal population density studies. There are also key confounding factors, including the initial radiation damage which damaged habitats in the most contaminated areas, and the major continuing ecosystem changes brought about in the Exclusion Zone due to the removal of the human population and the cessation of associated agricultural, forestry, hunting and fishing activities.

3. Discussion

Research following the Chernobyl accident resulted in considerable advances in radioecological research some, but not all, of which we have highlighted above. For a period the accident also prompted wider studies to improve our radioecology understanding in the event of future nuclear accidents.

It has been predicted that remediation measures will be required in inhabited settlements of the fSU until at least 2045 (Fesenko et al., 2007) and restrictions still remain in Scandinavian animal production systems. It is not surprising therefore, that high amongst the post-Chernobyl advances were the development of effective remediation approaches for long-term contaminated ecosystems. However, there was also realisation that the remediation strategies should not simply be focussed on the most scientifically and cost effective measures but should also take account of wider social factors, e.g. influencing acceptability (Lochard, 2007; Howard et al., 2005; Fesenko et al., 2007) with stakeholder groups being established in some countries to help plan acceptable remediation strategies (Nisbet et al., 2005). It could be argued that a consequence of the Chernobyl accident was to give radioecologists a stronger societal role.

Advances in the understanding of factors influencing the availability of radioaeasium from soils led to the development of spatially applicable models based on soil properties (e.g. Gillett et al., 2001; Wright et al., 2003) some of which were designed to aid in the selection of remediation approaches (Cox et al., 2005). Advances in, and the affordability of, GPS and mobile technologies gives the possibility of using geographical positions to predict 137Cs concentrations in free-ranging livestock such as reindeer (Skuterud et al., 2014) giving the potential for new management options to this long-lasting challenge.

After 30 years, the effects of radiation on organisms at Chernobyl are still not clear. If observations of major population-level damage on organisms at dose rates as low as a few μSv h⁻¹ are true, this has serious implications for our understanding of radiation protection of both humans and wildlife. There has been little international concerted effort to conduct radiation studies at Chernobyl. Apart from many studies by Ukrainian, Belarusian and Russian scientists in the early years (mostly published in the Russian language literature), there has been little concerted effort to conduct radiation studies at Chernobyl. If this issue is seriously to be addressed, more independent studies of radiation effects at Chernobyl (and Fukushima) are needed, and these studies must make great efforts to rule out potential sources of confounding and bias in experimental design and data analysis (Chesser and Baker, 2006).

The 30th anniversary of the Chernobyl accident occurs in the same year as the 5th anniversary of the Fukushima accident. The consequences of Fukushima, with evacuation of populations and consequences for farmers and producers again highlights the human dimensions of radiological accidents. In the modern society with social media etc. this is even more visible and evident than after Chernobyl and calls for radioecologists to take a more active role in society. Hopefully the knowledge gained after the Chernobyl accident will be of value in dealing with the Fukushima accident consequences though local factors, including societal, will influence how locally applicable such knowledge is. For instance, models of Cs availability based on soil properties (e.g. Absalom et al., 1999) do not appear to be working well for Japanese soils (Uematsu et al., 2016), whilst a greater focus of the remediation effort in Japan has been directed to reducing external exposure (IAEA, 2015). Using caesium binders as feed additives is an example of a measure (successfully used after the Chernobyl accident) which could improve farmers’ self-support and the utilisation of local feed resources. A positive reaction following the Chernobyl accident in Europe was the funding of multinational radioecological research programmes including collaboration between the affected fSU states and western Europe (e.g. Howard and Desmet, 1998). To date, no such international initiatives have been proposed with respect to the Fukushima accident.

Dedication

We dedicate this paper to the memory of all of those scientist we have worked with following the Chernobyl accident who are unfortunately no longer with us, including: Stuart Allen (UK); Nikolay Arkhipov (Ukraine); Panayotis Assimakopoulos (Greece); Petr Bondar (Ukraine); Vladimir Borzilov (Russia); Anatoly Bulgakov (Russia); Phil Day (UK); Slava Firsakova (Belarus); Rudie Heling (Netherlands); Nizam Isamov (Russia); Yuri Izrael (Russia); Jacob Kenigsberg (Belarus); Stanislav Kruglov (Russia); Vyacheslav Kyr-innyi (Ukraine); Nikolay Lochilov (Ukraine); Alexander Nikitin (Russia); Øyvind Pedersen (Norway); Alexander Petrovich (Belarus); Evgeniy Petrov (Belarus); Gennady Polikarpov

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