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Distillate ethanol production for re-use of abandoned lands - an analysis and risk assessment.

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Summary

Following the 1986 Chernobyl and 2011 Fukushima Daiichi nuclear accidents, large areas of land became unsuitable for crop production as a result of radioactive contamination. It is well known that distillation of fermented crops to produce ethanol significantly reduces impurities. This working paper presents the results of an experimental field study in the Chernobyl Exclusion Zone (CEZ) to evaluate the transfer of radionuclides to crops and to distilled ethanol. The Opachichi field site has contamination levels typical of the outer 10-30 km Zone and is significantly lower than many areas within the 10 km Zone. The ethanol is diluted to 40% by volume using water from the deep aquifer in Chernobyl town, 10 km south of the nuclear power plant. The rye grain had elevated levels of ^{137}Cs and ^{90}Sr, but Pu and Am isotopes were below detection limits. The ^{90}Sr activity was slightly above the Ukrainian limit of 20 Bq kg^{-1}. At this site within the CEZ, ^{90}Sr fallout is relatively high at 20 kBq m^{-2} and is much higher than in abandoned lands outside the CEZ. There were no artificial radionuclides observed in the distillate ethanol (diluted to 40% with Chernobyl Town groundwater) sample. The low energy beta analysis recorded an estimated 58 Bq/L which we attribute to natural ^{14}C consistent with the expected activity concentration of natural ^{14}C in ethanol at this dilution. All radionuclides analysed in the groundwater sample were below limits of detection. Modelling of radiation doses to a farm worker using three different models found these to be significantly below 1 mSv y^{-1}.

Introduction

Following the 1986 Chernobyl and 2011 Fukushima Daiichi nuclear accidents, large areas of land became unsuitable for crop production as a result of radioactive contamination. More than thirty years after Chernobyl, more than 6000 square kilometres of land in Belarus and Ukraine remain abandoned, a significant proportion of which are former agricultural lands. Since these accidents, many methods have been developed to reduce activity concentrations of radionuclides in crops (e.g. (Beresford et al., 2016; Fesenko et al., 2007), however in large parts of the abandoned areas, activity concentrations in crops could still exceed regulatory limits. Even in cases where regulatory limits (Japan: 100 Bq kg^{-1} of ^{137}Cs (Nihei et al., 2016); Ukraine: 50 Bq kg^{-1} ^{137}Cs, 20 Bq kg^{-1} ^{90}Sr for cereals (Balonov et al., 2018)) are not exceeded, there may be significant public reluctance to consume products viewed as "contaminated". For example, rice grown in the more contaminated parts of the Fukushima Prefecture has been traded at an approximately 20% lower price than the national average (MAFF, 2016) due to consumer concerns over its safety.

Alternative uses of crops grown in radioactively contaminated land have been suggested, including use as feed for fur-producing animals (Howard, 1993) and processing to produce industrial ethanol (Firsakova et al., 2000), both of which are expected to prevent the transfer of significant radioactivity to the human food chain. A stakeholder consultation in five Western European countries concluded that "any process that produces marketable food from contaminated raw materials was considered to be unacceptable" by the UK and Belgian stakeholder groups but may be acceptable under specific circumstances in some countries (Nisbet et al., 2005). This study (Nisbet et al., 2005) found that the use of contaminated crops for biofuel production was likely to be acceptable.

Any countermeasure based on processing of crops which exceed regulatory limits (or are deemed unacceptable by consumers for direct consumption) needs to be supported by a full life-cycle analysis of radiation risks to farm and process workers and evaluation of the fate of radioactivity in the original crop. The present study, for the first time, carries out such an analysis for the production of distillate ethanol from grain grown on experimental plots in the Chernobyl Exclusion Zone (CEZ) in Ukraine. Deep aquifer groundwater from the well in the town of Chernobyl (approximately 10 km South of the power plant site) is used for final dilution of the product to 40% ethanol by volume.

Methods

An experimental plot of approximately 0.25 ha was designated near the Opachichi settlement in one of the relatively less contaminated parts of the CEZ (Fig 1). The area is officially abandoned, but a few "self-settlers" remain. The soil type in the area is soddy-podzolic. Surface contamination of the field plot was 100 kBq m^{-2} of ^{137}Cs and 20 kBq m^{-2} of ^{90}Sr. Other isotopes were estimated from empirical data on isotope ratios to ^{137}Cs in this area (Chernobyl ECOCENTER; unpubl. res.). A crop of rye was grown on the field plot and harvested using standard farming methods.

Water samples were taken from a depth of > 250 m from a confined chalk-limestone aquifer of Jurassic age, the third aquifer from the surface in a multi-aquifer system, and the main source of urban water supply in Chernobyl town (Dzhepo and Skal'skii, 2002). The surficial Quaternary sandy alluvial unconfined aquifer (ca. 20 m thickness) received small amounts of radioactivity from surface fallout after the accident. But the deep aquifer is separated from the near-surface shallow groundwaters by another confined aquifer system of ca. >100 m thickness sandwiched by low permeability layers of marl and clay-rich lithologies at the top and bottom. Although there is spatial variability in the aquifer geology on a regional scale, this third aquifer, where the water sample came from, is isolated from the influence of the accident because of multi-level low-permeability barriers in between this and shallower unconfined aquifer (Fabyshenko and Nicholoson, 2015), as confirmed by previous tracer studies (Bugai et al. 1996).

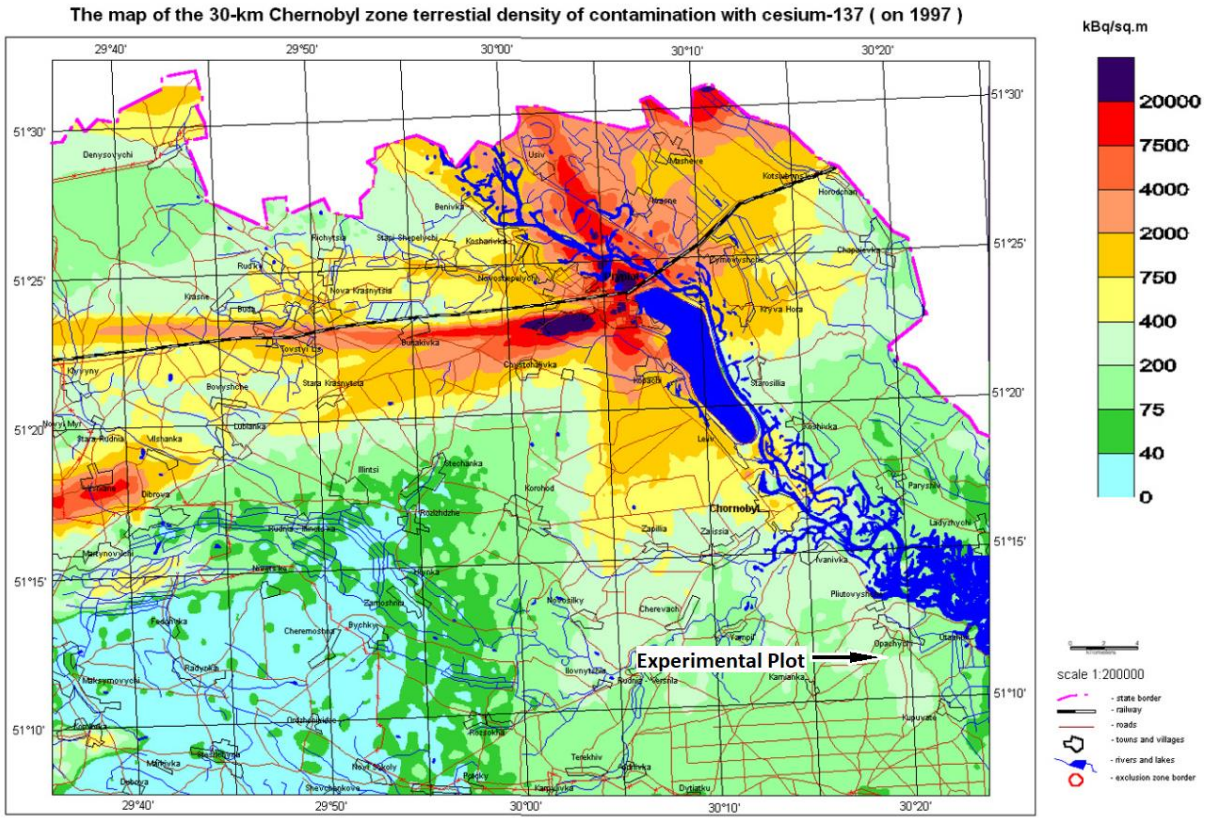

Figure 1. Map of contamination in the CEZ (as of 1997) and location of the experimental plot.

Distillation method

Ethanol production from grains involves a chain of different technical steps before the distillation process begins (Buglass, 2011). After harvesting, the grains were sieved to remove stones, stem and leaf residuals and other particles, and washed to remove dust. The grain is dried at 25-30 ^{o}C to reach relative humidity less than 50%, with further drying in VENTICELL forced air circulation heating oven at 45 ^{o}C to reach less than 15% humidity. The grain was then milled to a grain size of 0.2-1 mm with a flour fraction of less than 10%.

The wort was prepared in a 50 L experimental tank. 24 L of water was raised to a temperature of 70-80 ^{o}C then 6 kg of grain was added and stirred until it had the consistency of dense porridge. Alpha-amylase and glucoamylase were added to the Wort to liquefy and saccharify the starch molecules for further consumption by yeasts. A temperature range of 70-80 ^{o}C was used for the alpha-amylase stage for greatest efficiency of enzymatic breakdown of the starch. The glucoamylase is then applied when wort is cooled to a temperature of 60 ^{o}C (Balcerek et al. 2016). The wort was periodically mixed during the first 30 minutes after the enzyme was added. The wort was then left to be saccharified and cooled to 30 degrees prior to yeast fermentation (Neves et al., 2006).

Commercial active dried yeasts were used for fermentation at a proportion of 10 g of yeast per 1 kilogram of grain material. Fermentation was done in controlled temperature conditions over 5 days, for this purposes hand made insulated box with hot plate was used. Thermometer and controller kept the temperature of the wort at 30 ^{o}C during the full period of fermentation (Naeem et al., 2015). During fermentation, the wort was in a sealed reservoir with a tube for gas release with a water lock.

When the fermentation process is complete, the alcohol content in the wort is about 12%. The liquid part is decanted and filtered to remove suspended particles. From this time the liquid decantant of the wort (the “wash”) is ready for distillation. The distillation apparatus (AquaGradus Compact) with a 35 L stainless steel tank was used for triple distillation of the decanted liquid. The first distillation was carried out to obtain all raw alcohol without discrimination of spirits fractions, the temperatures of distillation of the first distillation were from 70 to 97 ^{o}C, and the distillation process was finished when the content of alcohol in the distillate flow decreased to 35%. Before the second distillation, all raw alcohol material from the first fraction was diluted with water to a content of 20% of alcohol in total. During the second distillation 10% of distillate was removed to "heads" fraction which correspond to the temperatures below 78 ^{o}C. The "Hearts" fraction were collected in a temperature range from 78 to 85 ^{o}C on low heat until the alcohol content in distillate flow decreased to 40%. The residuals of alcohol from the second distillation was discarded as the "tails" fraction that contain most of the fusel oils. Before the third distillation, the "hearts" from the second distillation were diluted with water to a content of 20% of alcohol in total and filtered through birch activated charcoal. During the third distillation, 5% of distillate was removed to the "heads" fraction. The "hearts" fraction were collected in a temperature range from 78 to 85 ^{o}C on low heat until the alcohol content in distillate flow decreased to 40%. The "hearts" from the third distillation was diluted with Chernobyl ground water to produce grain spirit with alcohol content 40%.

Radioanalytical methods

Radioanalytical methods at GAU-Radioanalytical were as follows:

Sub-samples of rye, diluted ethanol and groundwater were transferred to counting vials for analysis as received by gamma spectrometry. Further sub-samples were taken for ^{3}H, ^{14}C, gross alpha/beta analysis and digestion for all other radiochemical analysis.

Gross alpha/beta analysis: the rye sample was treated with sodium sulphate solution, ignited at 400^{o}C and then digested with aqua regia. An aliquot of the ethanol solution was dropwise applied to a filter and evaporated to dryness under an infrared lamp.

Radiochemical analysis: Sub-samples were spiked with ^{85}Sr, ^{232}U, ^{242}Pu and ^{243}Am tracers for chemical recovery monitoring. The spiked groundwater sub-sample was acidified with c.HNO_3 and evaporated to dryness and the residue digested with aqua regia. The rye sub-sample was ignited at 500^{o}C for >10hrs and the residue digested with aqua regia. The spiked ethanol sub-sample was evaporated to dryness and the residue digested with aqua regia.

Gamma spectrometry. High-resolution gamma spectrometric analysis was performed using HPGe detectors. Detectors were calibrated against a mixed radionuclide standard solution. The standard was used to prepare a source of identical geometry to that of the samples. Gamma spectra were analysed and individual radionuclides quantified using Fitzpeaks spectral deconvolution software (JF Computing Services). All artificial gamma-emitting radionuclides detected have been reported.

Screen by liquid scintillation counting. An aliquot of the sample was spiked into a vial containing a liquid scintillation cocktail and measured using a Quantulus ultra-low level liquid scintillation counter.

Gross alpha/beta in aqueous samples. The groundwater sample and prepared rye digest solution were further acidified with c.H_2SO_4 and the solutions evaporated to dryness. The resulting residue was ignited at 350°C. A sub-sample of the ignited residue was ground and mounted on a planchet and the source was counted by gas flow proportional counting. The counter was calibrated against ^{241}Am (alpha) & ^{137}Cs (beta).

Total tritium and ^{14}C. An aliquot of the sample was progressively combusted to 900°C in a silica work tube using air/O2 combustion / carrier gas. The combustion products were passed over a Pt-alumina catalyst and heated to 800°C to ensure the complete conversion of tritiated species to tritiated water and ^{14}C to $^{14}CO_2$. The tritiated water was then trapped in HNO_3 bubblers and the ^{14}C in Carbontrap bubblers. The $^{3}H/^{14}C$ collected in the bubblers was measured using a Quantulus ultra-low level liquid scintillation counter.

^{90}Sr in aqueous samples. The strontium was pre-concentrated by precipitation as an oxalate and purified using extraction chromatography. ^{90}Sr activity was determined by measuring the in-growth of the ^{90}Y daughter using Cerenkov counting. Chemical recovery was determined via measurement of the ^{85}Sr yield monitor in the final purified strontium fraction.

U by alpha spectrometry. A combination of anion exchange and extraction chromatography was used to isolate U from the prepared solution. The U was electrodeposited onto a stainless steel disc and ^{234}U, $^{235+236}U$ and ^{238}U activities determined by alpha spectrometry.

Pu by alpha spectrometry. The Pu was isolated from the prepared solution by anion exchange chromatography. The Pu was electrodeposited onto a stainless steel disc and the ^{238}Pu and $^{239,\ 240}Pu$ activities determined by alpha spectrometry.

^{241}Am by alpha spectrometry. The Am was isolated from the prepared solution by anion exchange chromatography. The Am was electrodeposited onto a stainless steel disc and ^{241}Am activity determined by alpha spectrometry.

Limits of Detection. Limits of detection for radiochemical analyses are quoted as defined by Currie, 1968. Limits of detection for gamma spectrometric analysis are quoted as defined by Gilmore and Hemingway, 2000. Limits of detection for alpha spectrometric analysis are quoted as defined by Hurtgen et al, 2000.

Table 1. Parameter values and distributions for Monte Carlo estimation of external and inhalation effective dose rates. Ratios of Sr, Pu, Am to ^{137}Cs are representative of the relatively less contaminated parts of the CEZ and would be higher at higher contamination densities.

Parameter	**Central estimate**	**Assumed variability**	**Notes**
$^{90}Sr/^{137}Cs$ ratio	0.32	Lognormal	ECOCENTER data
$LOG_{10}(^{90}Sr:^{137}Cs)$	-0.56	Lognormal (S.D. 0.25)	ECOCENTER data
$^{238}Pu/^{137}Cs$ ratio	2.62×10^{-3}	Uniform (1.54×10^{-3}-3.58×10^{-3})	ECOCENTER data
$^{239,240}Pu/^{137}Cs$ ratio	6.7×10^{-3}	Uniform (3.85×10^{-3}-9.17×10^{-3})	ECOCENTER data
$^{241}Am/^{137}Cs$ ratio	1.45×10^{-2}	Uniform (1.15×10^{-2}-15.8×10^{-2})	ECOCENTER data
Soil dry bulk density	1400 kg m^{-3}	Normal (S.D. 200)	Range from clay to sandy soils
Plough mixed depth	0.25 m	Uniform (0.2 – 0.3)	Typical range
Ploughed field dose	0.5 µSv h^{-1} per MBq m^{-2}	Uniform (0.35-0.7)	Empirical data (UIAR)
Occupancy farm worker	4.6 h ha^{-1} $year^{-1}$	Uniform (3.1 – 6.9)	(Williams et al., 2006)
Adult breathing rate (activity level "light")	0.86 m^3 h^{-1}	Normal (S.D. 0.15)	(Moya et al., 2011)
Inhalable dust soil tillage	2×10^{-5} kg m^{-3}	Uniform (5×10^{-6}-4×10^{-5})	(Arslan and Aybek, 2012)

Radiological risk assessment

The radiological risk assessment for distillate ethanol production is based on estimation of dose to a farmer/farm worker carrying out all soil preparation, crop spraying and harvesting operations across a 100 ha farm, a reasonable estimate farm size for one farmer/farm worker. A Monte Carlo approach (1000 model runs) was taken to evaluating variability in external dose rate, using estimated parameter ranges and distributions. It was assumed that field operations (soil preparation, seeding, spraying and harvesting) took 4.6 hours/ha, a conservative estimate (based on data for the UK in (Williams et al., 2006)), reflecting potential use of smaller farm machinery than is typical in the UK. Potential variation was assumed to be a factor of 1.5 above or below this value, with a uniform distribution between upper and lower limits. External exposure is estimated for a soil activity-depth profile corresponding to a mixed (ploughed) depth of 0.25 m, a soil density of 1400 kg m^{-3} and 100 kBq m^{-2} surface ^{137}Cs density. The assumed dose conversion coefficient for a ploughed field was 0.5 µSv h^{-1} per MBq m^{-2} based on empirical data (Ukrainan Inst. of Agricultural Radiology, unpubl. res.). Inhalation dose coefficients were taken from (IAEA, 2004a). Parameters used and assumed probability distributions are summarised in Table 1.

Results and Discussion

Radionuclide isotope ratios

The ratios of ^{90}Sr, ^{241}Am and isotopes of Pu in fallout are presented in Table 2. Measured ratios in this part of the CEZ are 3.5-4.0 times higher than those in the Chernobyl release due to preferential fallout of these less volatile radionuclides closer to the accident site (Mück et al., 2002). These are representative of relatively less contaminated areas of the CEZ (the majority), but would be higher for more contaminated areas (for example, the ^{90}Sr:^{137}Cs ratio approaches 1 in the most contaminated "Red Forest" area). It can be seen that (even accounting for further future ingrowth of ^{241}Am from ^{241}Pu), contamination densities of transuranium elements are low compared to ^{137}Cs and ^{90}Sr.

Table 2. Radionuclide ratios to ^{137}Cs in the study region of the CEZ.

Radionuclide ratio	N	Measured ± S.E.	Chernobyl release*
^{90}Sr:^{137}Cs	15	0.32 ± 0.048	0.12
^{238}Pu:^{137}Cs	9	$2.6\ (\pm 0.24) \times 10^{-3}$	6.7×10^{-4}
$^{239+240}Pu$:^{137}Cs	9	$6.7\ (\pm 0.65) \times 10^{-3}$	1.8×10^{-3}
^{241}Am:^{137}Cs	9	$1.4\ (\pm 0.04) \times 10^{-2}$	3.9×10^{-3} **

* (UNSCEAR, 2000a); ratios are decay corrected to 2018. ** Including 32 years' ingrowth from Pu-241.

Activity concentrations in grain

As shown in Table 3, the rye grain had elevated levels of ^{137}Cs and ^{90}Sr; the latter being slightly above the Ukrainian limit of 20 Bq/kg. At this site within the CEZ, ^{90}Sr fallout is relatively high at 20 kBq m^{-2}. Outside the CEZ in Zone 2 ("Zone of Obligatory Resettlement") ^{90}Sr fallout is much lower so, for the same ^{137}Cs contamination level, ^{90}Sr activity concentrations in crops are likely to be lower than the Ukrainian limit. ^{14}C and ^{40}K activity concentrations in the rye

were similar to those expected from levels of natural radioactivity in crops (IRSN, 2012), so no evidence of ^{14}C fallout from Chernobyl was seen. The ^{3}H level of 100 Bq kg^{-1}, though of no radiological significance due to the low beta energy of this isotope, was unexpected as it is significantly higher than expected natural ^{3}H and requires further investigation to determine whether it is due to ^{3}H of Chernobyl origin. ^{241}Am and isotopes of Pu were all below the limit of detection.

Table 3. Radioactivity in ground rye grain (Bq/kg d.w.).

Analysis	Activity concentration	Notes
Gross alpha	< 20	
Gross beta	250 +/- 30	Mainly natural ^{40}K with some ^{90}Sr. Does not include low energy betas.
^{3}H (Tritium)	100 +/- 50	Above expected natural background, but of very minor radiological significance. No evidence of ^{3}H in the ethanol sample.
^{14}C	60 +/- 30	Within the range of expected natural background.
^{90}Sr	26 +/- 8	Radioactivity from Chernobyl and slightly above the Ukrainian limit for grain (20 Bq/kg).
^{137}Cs	2 +/- 1	Radioactivity from Chernobyl and below the Ukrainian limit for grain (50 Bq/kg).
^{60}Co	< 3	
^{241}Am	< 0.2	
^{234}U; $^{235+236}U$; ^{238}U	< 0.08; <0.06; < 0.06	
^{238}Pu; $^{239+240}Pu$	< 0.07; < 0.1	
^{40}K	140 +/- 30	Natural radioactivity.
^{228}Ac; ^{212}Pb; ^{212}Bi; ^{208}Tl; ^{235}U; ^{234}Th; ^{226}Ra; ^{214}Pb; ^{214}Pb; ^{210}Pb	All below limit of detection	Natural radioactivity.

Radioactivity concentrations in distillate ethanol

There were no artificial radionuclides observed in the distillate ethanol (diluted to 40% with Chernobyl Town groundwater) sample (Table 4). The low energy beta analysis recorded an estimated 58 Bq/L which we attribute to natural ^{14}C consistent with the expected activity concentration of natural ^{14}C in ethanol at this dilution.

Table 4. Radioactivity in 40% distillate ethanol (Bq L^{-1}).

Analysis	Activity concentration	Notes
Gross alpha	< 7	
Gross beta	< 10	
Alpha; liquid scintillation	< 4	
Low energy beta	58 +/- 0.5	Detects ^{14}C: this activity is consistent with expected natural ^{14}C in ethanol
High energy beta	< 10	
^{90}Sr	< 0.6	
^{137}Cs	< 1	
^{60}Co	< 3	
^{241}Am	< 0.02	
^{234}U; $^{235+236}U$; ^{238}U	< 0.006; <0.008; < 0.005	
^{238}Pu; $^{239+240}Pu$	< 0.009; < 0.007	
^{40}K	< 40	
^{228}Ac; ^{212}Pb; ^{212}Bi; ^{208}Tl; ^{235}U; ^{234}Th; ^{226}Ra; ^{214}Pb; ^{214}Pb; ^{210}Pb	All below limit of detection	Natural radionuclides

Radioactivity concentrations in water

All radionuclides analysed in the groundwater sample were below limits of detection as shown in Table 5.

Table 5. Radioactivity in deep groundwater - Chernobyl Town (Bq L^{-1}).

Analysis	Activity concentration	Notes
Gross alpha	< 0.7	
Gross beta	< 1	
Alpha; liquid scintillation	< 5	
Low energy beta	< 4	Detects ^{14}C: this lower limit is consistent with natural ^{14}C in very old groundwater.
High energy beta	< 3	
^{90}Sr	< 0.1	
^{137}Cs	< 1	
^{60}Co	< 2	
^{241}Am	< 0.0003	
^{234}U; $^{235+236}U$; ^{238}U	< 0.001; <0.001; < 0.001	
^{238}Pu; $^{239+240}Pu$	< 0.001; < 0.001	
^{228}Ac; ^{212}Pb; ^{212}Bi; ^{208}Tl; ^{235}U; ^{234}Th; ^{226}Ra; ^{214}Pb; ^{214}Pb; ^{210}Pb	All below limit of detection	Natural radionuclides

Dose assessment to agricultural and process-workers

The estimated range of external dose rates to an agricultural worker using generally conservative assumptions (e.g. no shielding from tractor or combine harvester, relatively high exposure time) is given in Figure 2. External dose rate is shown for 100 kBq m^{-2} of ^{137}Cs contamination density. Note that dose to the agricultural worker is only for time spent on a ploughed field (and is additional to natural background) whereas natural external dose is for full year exposure.

Figure 3 shows the range of effective dose rates from inhalation, primarily from alpha-emitting isotopes of Pu and ^{241}Am. Doses are estimated for an agricultural worker at two different contamination densities of ^{137}Cs. It is assumed that she or he is working without the protection of a cab on a tractor or combine (which would, if present, significantly reduce inhalation of fine particulates). Even at high inhalable dust levels, doses from Chernobyl-derived radioactivity are small compared to the range of doses worldwide from natural alpha-emitting ^{222}Rn and its progeny.

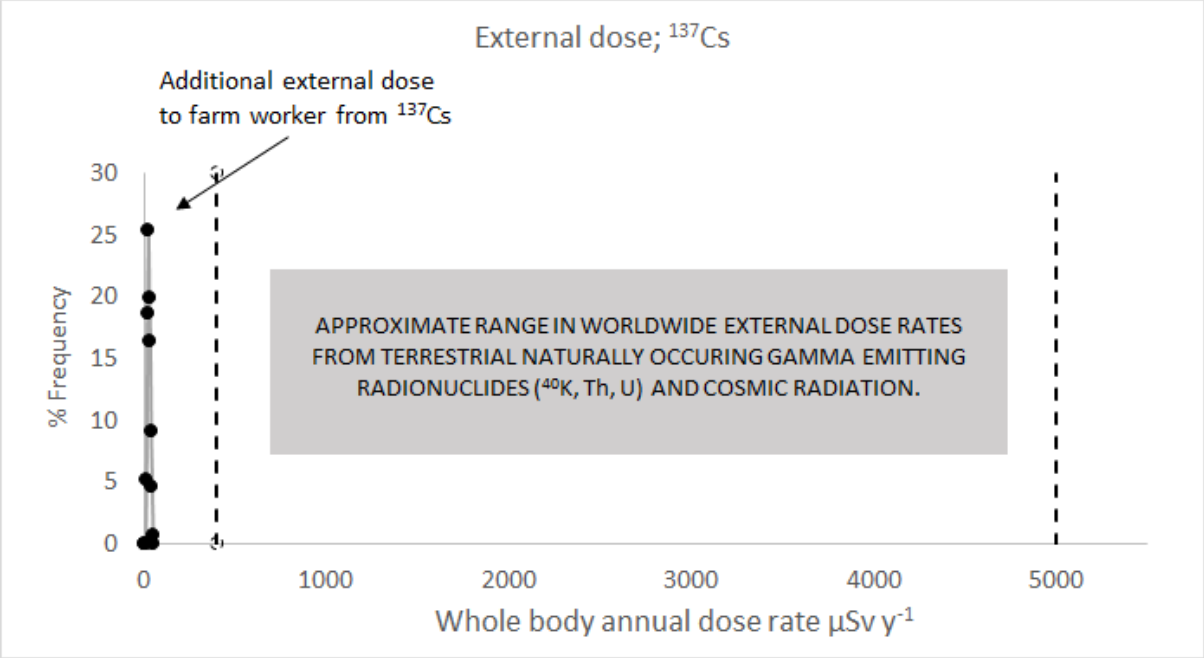

Figure 2. Range in effective annual gamma dose rate to an agricultural worker at the field study site compared to illustrative range in annual external dose rates worldwide from naturally occurring terrestrial gamma emitters (UNSCEAR, 2000a) assuming a conversion from absorbed dose in air to external effective dose of 0.7 Sv Gy^{-1} (UNSCEAR, 2000b) and cosmic radiation (Bouville and Lovett, 1988). Natural terrestrial external dose rates in Northern Ukraine are at the lower end of this range.

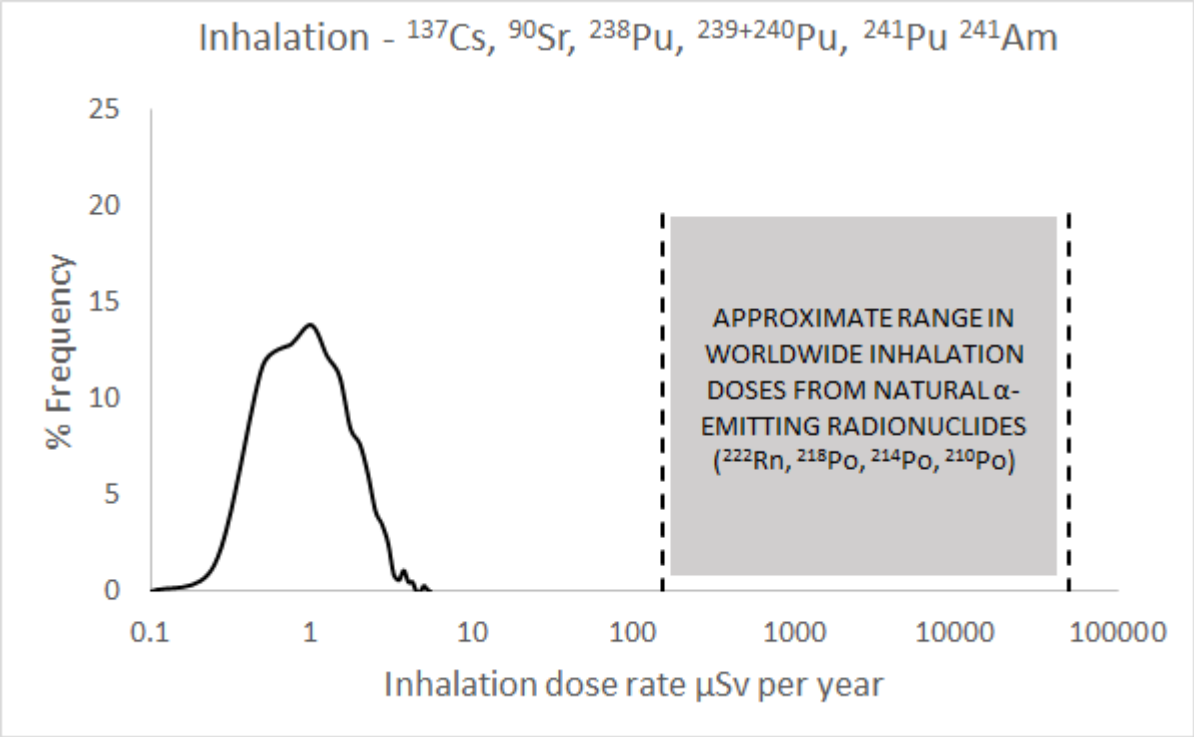

Figure 3. Range in total (^{137}Cs, ^{90}Sr and alpha-emitters) effective equivalent inhalation dose rates from agricultural activity at the experimental site (100kBq m^{-2}) ^{137}Cs contamination density in the CEZ. The illustrative range of effective equivalent dose rates worldwide from natural alpha-emitting radionuclides is also shown (based on data in (Appleton, 2007; Dubois, 2005)).

Effective equivalent doses from external, inhalation and inadvertent ingestion of soil were also calculated using the RESRAD (Yu et al. 2007) and NORMALYSA (Avila et al., 2018) software. Mean external dose was 13.4 μSv y^{-1} in both models and inhalation dose was 3.4 and 11.8 μSv y^{-1} respectively. These are broadly consistent with doses estimated in the MC model, though external dose and hence total dose is lower due to a lower ploughed field soil-external dose rate coefficient assumed in the RESRAD and NORMALYSA models. Effective equivalent dose rates from inadvertent soil ingestion were insignificant in comparison to external dose. The doses to a farm worker are therefore well below the reference occupational (non-classified worker) dose rates.

Dose assessment for consumers

There is no evidence of any significant dose to consumers above natural background.

Conclusion

Levels of radionuclides of Chernobyl origin are slightly elevated in rye grain grown at the field study site. Cs-137 is significantly below the Ukrainian regulatory limit, but ^{90}Sr, at 26 +/- 8 Bq kg^{-1} is slightly above the 20 Bq kg^{-1} limit. As expected, distillation of the grain to produce distillate alcohol reduced radioactivity (including natural ^{40}K) very significantly such that no radioactivity except for natural ^{14}C was found in the distillate. Groundwater at Chernobyl town, used to dilute the distillate, was, as expected, free from artificial radioactivity. The doses to a farm worker are therefore well below the reference occupational (non-classified worker) dose rates.

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