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Decontamination after a nuclear fallout: A condensed review of case studies, methods and key references up to 2014

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Abstract and objectives

The objective of this work is to provide governmental agencies and the society a condensed overview of the current state of knowledge on the efficiency of applied restoration and remediation of areas affected by radioactive fallout. The most actual real-world case is the Fukushima accident in Japan in 2011: the accident and the following applied decontamination methods are in focus in this review study. This report was written in 2014 and reviewed and translated in 2018/2019. This work was financed by the Swedish Civil Contingencies Agency.

Introduction

An exercise held at Samö, Sweden, 2011 (Swedish Civil Contingencies Agency, 2011) focused on the societal restoration after a radiological accident. The outcome of the exercise showed that Sweden has a great need of improving its knowledge and development with regards to restoration and remediation of the society following a nuclear fallout. Simultaneously as the exercise took place in Sweden, the Fukushima accident occurred in Japan. Studies and investigations on the management, restoration and remediation of the nuclear fallout in Japan are of great interest in terms of transferring knowledge from a real-world situation into a context of nuclear-/radiological emergency preparedness. The experiences from Japan is an important addition to the already existing knowledge that has been generated from experiences of the Chernobyl catastrophe in 1986 and through field exercises.

Regarding the remediation in Japan there is an issue of imprecise description of the methods used. Additionally, the areas that per 2014 have finalized any decontamination program are relatively few. Hence, data on achieved dose reduction is scarce. It has been shown that the decontamination efficiency in Tamura city reached 25% (Ministry of Environment, 2014). It is however still unclear why only a 25% dose reduction in this area could be reached. Other locations, such as parks, schools and houses in Japan have reached a mean decontamination efficiency of 56%, however with a large span of variation (9-78%) (Ministry of Environment, 2014). The definition of decontamination efficiency and a calculation example is given in Appendix 1.

The main goal with this overview is to find and explore contemporary (by 2014) effective methods for decontamination and restoration of radioactively contaminated areas. It is likely that this type of survey still is too close in time after the Fukushima accident to find the same amount of relevant literature as there are for the Chernobyl accident. Hence, full implementation of the knowledge and experiences from the Fukushima decontamination process will probably not be fully reported in methodological literature on decontamination and restoration until after a number of years. In this report we describe some of the applied decontamination methods and their efficiency.

Information paths to literature on decontamination of radioactive fallout

Experiences from decontamination and related research following actual radiological and nuclear accidents, such as the Chernobyl release in 1986, have generated several innovations in forms of methods, techniques and products that are designed to facilitate decontamination. A thorough review of decontamination methods can be

found in the European approach to nuclear and radiological emergency management and rehabilitation strategies (EURANOS) handbook (Nisbet, 2011). The EURANOS handbook contains and describes 59 different decontamination strategies. The handbook is crucial for decision makers and other professionals that are working with decontamination and radiological-/nuclear emergency preparedness.

Fukushima accident March 2011 – decontamination progress

The most straight forward approach to get information regarding the planning and performance of applied decontamination actions in Japan after the Fukushima accident is through the Japanese Ministry of Environment (MOE) webpage (<http://josen.env.go.jp/en/>). The webpage is continuously updated. Except from updates regarding the decontamination process, one can also find information about the following topics:

- Policy Framework
- Progress in the Special Decontamination Area
- Progress in the Intensive Contamination Survey Area
- Decontamination technology development
- Efforts to secure the Interim Storage Facility

Contaminated municipalities were divided into two subcategories, “Special Decontamination Area” (SDA) and “Intensive Contamination Survey Area” (ICSA). The Japanese state is responsible for the decontamination in the “Special Decontamination Area” until all evacuated former residents are able to return to their homes. The area is, at the time of writing, currently containing 11 municipalities and the yearly dose prior decontamination exceeds 20 mSv. The “Intensive Contamination Survey Area” consist of 100 municipalities and the municipalities are responsible for the decontamination. This area is characterized of the air dose rate exceeding $0.23 \mu\text{Sv/h}$. In August 2013, the SDA was classified into three areas according to Table 1.

Table 1. Categorization of the contaminated areas in the vicinity of Fukushima, Japan.

Green area	1)	<20 mSv/yr	Evacuation orders are ready to be lifted
Yellow area	2)	20-50 mSv/yr	Residents are not permitted
Red area	3)	>50 mSv/yr	Residents will have difficulties in returning for a long time

The distribution of results from around 250 000 measurements (prior and post decontamination, by 2014) of the dose rate in air shows that the 25-percentile was $0.36 \mu\text{Sv/h}$ prior and $0.25 \mu\text{Sv/h}$ post decontamination. The 75-percentile value was $0.93 \mu\text{Sv/h}$ prior and $0.57 \mu\text{Sv/h}$ post decontamination.

The total volume of radioactive waste generated through decontamination was estimated to $15 \cdot 10^6$ to $28 \cdot 10^6 \text{ m}^3$ (data from 2014). There are obvious issues in finding appropriate storage for this amount of waste.

Decontamination areas

The decontamination efficiency achieved is not only dependent on the method and equipment used but also in what environment remediation work is carried out. Decontamination areas after the Fukushima accident are usually categorized as:

- i.) Residential houses (inhabited areas)
- ii.) Farmland
- iii.) Forest close to residential area
- iv.) Roads

Cases studies of decontamination efficiencies

The need for remediation of the outdoor environments after a nuclear accident usually depends on ^{137}Cs , and in the early years also ^{134}Cs . The amount of caesium that can be cleaned and removed varies with location and applied methodology. What can be expected as a good remediation efficiency can be found in the EURANOS manual (Nisbet, 2011). Below we present six case studies to cover the typical and most common decontamination practices.

Case 1 – Bryansk, Russia, 1989

Decontamination in 1989 of 93 settlements with about 90,000 residents in Bryansk, made by the Russian Army (K.G. Andersson, 2003). The remediation included only two procedures:

1. Removal of the top surface layer (5-10 cm) in private gardens, around public buildings and along 190 km of roads.
2. As a protective shield from the remaining soil contamination, a layer (5-10 cm) of pure coarse sand was then laid out.

The decontamination procedures reduced the dose rate by between 9 and 30%. Same remediation technique was later applied in Kirov, Belarus. Here, the dose rate was decreased by 8%, the remedial process was continued for a number of years. The decontamination work was assessed as non-cost effective but sufficiently risk-eliminating. However, the possibility for residents to cultivate and grow in the remaining soil is greatly impaired.

Case 2 – Novo Bobovichi, Bryansk, Russia, 1995

During the autumn of 1995 the Centre for nuclear technology at Denmark Technical University in Risø, Denmark, sent a Danish-Russian group to decontaminate three wooden houses in Novo Bobovichi, north of Novozybkov, Russia (K.G. Andersson, 2003). Decontamination was done in the following procedure:

1. Removal of a 5-10 cm surface layer from a surface of 20x20 m adjacent to the houses with spades and shovels.
2. Clean the house's asbestos roof from the leaves, pine barrels and other litter, then clean the roof with a water spraying machine.
3. Place clean coarse sand over ground contaminated surfaces.

On average, the dose rate indoors was reduced by 64% after this work. Outside of the houses, the reduction was 78%.

Case 3 - Novo Bobovichi, Bryansk, Russia, 1997

The Danish-Russian group returned to Bobovichi in 1997 to decontaminate 5 houses (Roed, 1998; K.G. Andersson, 2003). This time, machines were used in the decontamination work as follows:

1. A Bobcat was used to put the soil layers in highs/lumps/piles.
2. A team of workers carrying dosimeters and shovels were sent out to look for and removing hot spots.
3. An excavator drove over to cleaned surfaces and dug deep pits for waste.
4. The Bobcat was used to move the waste to the pitfalls.
5. The clean sand from the pits were then placed by the Bobcat over the scratched surface and restored the previous ground level.

Case 4 – Halland, Sweden, 2006

During the exercise DEMOEX in the region of Halland, Sweden, two areas of grass-covered surfaces were contaminated by the short-lived nuclide $^{99}\text{Tc}^m$. Immediately after $^{99}\text{Tc}^m$ application the surface layers were removed by an excavator and effectively decreased the remaining activity of the two areas to 2 and 9,5 %, respectively (Figure 1; Finck, 2012). In this case no material with gamma-ray attenuating properties was distributed on top of the surfaces.



Figure 1. Surfaces around the house during the DEMOEX-exercise have been removed.

Case 5 – Tamura, Japan, 2011–2014

The decontamination process following the Fukushima accident is continuously monitored and evaluated. Many municipalities are already decontaminated to a sufficient level (Tamura, Naraha, Kawauchi, Okuma) at the time of writing this report (2014). Japanese decontamination teams are dominating the remediation labor, while many international groups are assisting with the measurements.

The report by Hiraoka et al. (2014) contain relevant data regarding decontaminated areas. Detailed information can be found for the decontamination process of the town Tamura. Tamura was decontaminated to sufficient levels in June 2013. Decontamination of the 5 km² area started in July 2012 and was completed in June 2013. The numbers of workers involved in the decontamination reached a maximum of 1 300 workers per day and resulted in a total labor input of approximately 120 000 person-days of labor. Spatial objects that were target for decontamination included 228 000 m² of buildings, 96 km of roads, 1 300 000 m² of farmland and 1 900 000 m² of forests.

The evacuation was ceased in Tamura in April 2014. Dose rates were measured prior and post decontamination in Tamura. The lower dose rate measured after the decontamination process can partly be explained by ¹³⁴Cs decay. The overall decontamination efficiency can be found in the interval of 21-36% and is as expected low compared to the decontamination efficiency that was achieved during the DEMOEX exercise (Case 4).

A very informative report “Overview of the Results of the Decontamination Model Projects” gives examples of decontamination projects in the aftermath of Fukushima (JAEC, 2012). The decontamination efficiency calculated from dose rate measurements (1 m above the ground) is given for several examples. The mean decontamination efficiency shows large variation between different objects, however it seldom exceeds 50%. The decontamination efficiency in Yonomori Park was described in the abovementioned report. The area consists of skyscrapers, smaller residential houses, streets with trees, playgrounds and a forested area. The dose rate prior decontamination was 7.9 μSv/h. Mean dose rate post decontamination was 4.2 μSv/h. Decontamination efficiency was determined to be 47%.

Pin-pointing factors that contribute to a lower decontamination efficiency is hard. It can be speculated that application of high pressurized water without collecting the residual contaminated water may be one factor that contributes to a lower decontamination efficiency. Imprudence in using potentially contaminated machines, equipment and soil material which causes secondary distribution of the radionuclides may be another aggravating factor.

Case 6 – Baseball field, Japan, 2011-2014

The Japanese Atomic Energy Agency, JAEA, decontaminated a baseball field of 4 800 m² as a research project on decontamination efficiency (Figure 2). Except from removing the top-soil layer and later adding a 5 cm layer of clean sand on the surface, other decontamination methods used in this case is not known to the authors of this reports. However, there are measurement data prior and post decontamination. A measurement team from Sweden visited the decontaminated baseball field in 2014 and presented the following results:

- The dose rate in the centrum of the baseball field was measured to be 26 µSv/h in March 2013, prior decontamination.
- The dose rate in the center of the baseball field was measured to be 1.6 µSv/h in November 2013, post decontamination.
- Decay correction applied to ¹³⁷Cs and ¹³⁴Cs gives a decontamination efficiency of >90%.



Figure 2. Decontaminated baseball field, Japan.

This case can be considered as a successful decontamination. According to the EURANOS handbook a dose rate reduction of a factor 5 to 10 is to be expected when applying decontamination methods on plane surfaces. However, there are some factors in this case that can be considered crucial for any decontamination result:

- A plane surface, like a baseball field is relatively easy to decontaminate. There is no vegetation that can act as obstacles during the decontamination.
- The added 5 cm of sand onto the surface is attenuating the gamma radiation from the buried radionuclides.
- The adjacent forest situated in the northern direction of the baseball field was not decontaminated (see Figure 2).

In Appendix 1 in this report one can find calculations showing how contaminated surfaces from various distances affect the dose rate in locations within a decontaminated area.

Waste management

The storage of the enormous amounts of contaminated soil and other types of waste that has been generated through the decontamination process in and around Fukushima creates some issues. While waiting for a solution for long-term storage the waste is temporarily stored in so called Interim Storage Facilities (ISF). The waste types stored in ISFs are contaminated soil, building material and ash, all material with an activity

exceeding 100 kBq/kg. There has been some struggle to achieve public acceptance for localization of these ISFs (MOE, 2014). It is probably likely that if a similar situation happened in Sweden, there will be some issue in finding relevant and appropriate locations for ISFs as well as reaching public acceptance of these types of storage.

Research – Current and emerging

The former Swedish agency, Räddningsverket, published their general advices in 2007 (Räddningsverket, 2007). Since then, scientific literature on nuclear decontamination in a Swedish context is very scarce.

Recent studies have shown potential to clean different materials from caesium with just water. A Japanese research group has shown in laboratory trials and an applied test in Fukushima that water-containing bubbles in the nano-size can act as a potential cleaning agent (Yoshikatsu Ueda, 2013). In 100 g of dried coarse gravel, 2% of the caesium could be extracted by clean water while the nano-bubble water could extract 17% of the caesium. This type of research results shows the potential existence of environmentally friendly decontamination techniques.

Conclusions

1. Large scale decontamination projects have been performed in Japan. The decontamination efficiency of the applied methods seems, in many cases, to be lower than what is described in decontamination handbooks (i.e. EURADOS). One reason can be that secondary distribution of caesium has occurred. In some highly monitored and controlled areas higher decontamination efficiency has been achieved, this may be due to well-tested methods and trained staff.
2. Large amounts of radioactive waste are produced in decontamination actions and the location for storage and achieving public acceptance are major issues.
3. Recent research studies show promising results in finding green technology applied for decontamination of radiocaesium. Research results from the decontamination process in Japan is expected to emerge greatly in the coming years.
4. Some decontamination methods need to be tested experimentally in Swedish conditions, i.e. the method dependence upon snow and ice cover.
5. Decontamination exercises using short-lived radionuclides are meaningful and instructive and would most likely elevate the state of knowledge in the field of nuclear decontamination. These kinds of experiments are, however, demanding high resources.
6. It is practically impossible to remove all radiocaesium from large areas. In order to achieve the best and most cost-effective decontamination effect a synthesis is needed to describe how different decontamination methods can be used together.
7. Nuclear accidents are very rare. When an accident occurs, the knowledge on decontamination may be totally forgotten. Easy access instructions and advices regarding decontamination in different environments are crucial. The EURANOS handbook is an essential information source in decontamination and provides a framework for restoration procedures.
8. An effective decontamination process requires radiometry trained staff. Hotspots of radiocaesium need to be identified, and if possible, removed.
9. A well-prepared decontamination strategy is a necessity in order to avoid secondary distribution and spread of radiocaesium.

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Appendix 1

Decontamination efficiency, E

The most used parameter for measuring the success of a decontamination action is called the decontamination efficiency, E . It shows the relative decrease of the gamma radiation net dose rate after a decontamination action. It is important to use the net dose rate if the background radiation is significant. In these cases, the natural background dose rate must be subtracted from the total dose rate measured before and after decontamination. As an example, if the gamma radiation dose rate is measured to be $0.90 \mu\text{Sv/h}$ prior decontamination and a decontamination action decreases the measured dose rate to $0.57 \mu\text{Sv/h}$, then assuming a natural background gamma dose rate of $0.10 \mu\text{Sv/h}$, the decontamination efficiency, E , can then be calculated as:

$$E = \frac{(0.90 - 0.10) - (0.57 - 0.10)}{(0.90 - 0.10)} \cdot 100 = \frac{0.80 - 0.47}{0.80} \cdot 100 = 41\%$$

Decontamination efficiency can also be expressed as the dose reducing factor, DRF . DRF is calculated as the ratio between the net dose rate after and prior decontamination. Applying the calculation of DRF on the numbers used in the example above gives:

$$DRF = \frac{0.47}{0.80} = 0.59$$

This gives us the relation between E and DRF to be:

$$E = (1 - DRF) \cdot 100$$

When decontamination actions are applied on surfaces such as walls, roofs and floors the physical quantity decontamination factor, DF , is often used. It describes the ratio of the activity per area unit (Bq/m^2) prior and after decontamination.

Decontamination of a limited surface in an area of widespread radioactive deposition

When a radionuclide is in target for decontamination actions on a specific surface or area through removal or radiation shielding, the effect is seldom a total removal of the radionuclide. A certain air dose rate is almost always still present. It is very hard to remove all of the radionuclides that are contaminating an area. Further, an area that is subjected to decontamination is a limited area, however gamma radiation has no determined range which means that adjacent non-decontaminated areas may still contribute to the dose rate in the decontaminated area. This scenario is always valid but more important in small decontaminated areas.

A simple theoretical model can show how gamma radiation from an adjacent contaminated area is contributing to the air dose rate in a decontaminated area. It is common to measure the air dose rate one meter above the ground. Assume successful (although unrealistic) decontamination, i.e. a total removal of all radionuclides in circular surface area. Figure A1 shows the decontamination efficiency for a nuclear fallout deposition containing radiocaesium as a function of decontamination radius for various penetration depths, using a soil model described in Beck et al. (1972). The efficiency is expressed as the relative decrease in air dose rate above a completely decontaminated circular surface with varying radius. If the area of the decontaminated area is small (a couple of meter radius) a large fraction of the measured air dose rate above the decontaminated area will origin from the surrounding contaminated area. The decontamination efficiency will in these types of cases be small (10–20%). In order to achieve a larger decremental effect on measured air dose rate the area subjected for decontamination must be increased to at least a few hundred m^2 . Around 10 meters radius in the decontamination field can result in a 40-80%

decontamination efficiency.

The curve fitting in Figure A1 can be used to estimate the effect on air dose rate from a remaining adjacent contaminated area. If a circular area of 10 meters in radius is decontaminated with a removal of 20% of the present radionuclides then the decontamination efficiency will decrease from 77% to $77 - (77 \times 20 / 100) = 62\%$, assuming a 3 cm depth mixing in the top soil.

The ground penetration of the radionuclide affects the measured air dose rate above the decontaminated area. If all radionuclides are deposited at the surface of the ground with no ground penetration, then air is the only attenuating media from the surrounding contaminated area. The attenuation coefficient of air is small, which results in high contribution of the dose rate from the adjacent contaminated area. However, the gamma ray attenuation will be significant if the radioactive fallout has penetrated the ground. The attenuation gets even stronger with the distance to the penetrated radionuclides since the radiation gets an oblique angle to the decontaminated area which means that larger amount of soil-/ground material needs to be passed and thereby a larger attenuation. Hence, the deeper radionuclide penetration, the larger shielding of gamma rays. Consequently, smaller areas may be subject to decontamination when deep ground penetration has occurred. As an example, to achieve a 50% reduction in air dose rate in a ^{137}Cs contaminated area with the radionuclide on the surface a circular area of 14 m radius needs to be decontaminated. However, if the ^{137}Cs has penetrated and is distributed within the top 3 cm of the ground, only a 4 m radius circular area needs to be decontaminated to achieve 50% dose rate reduction.

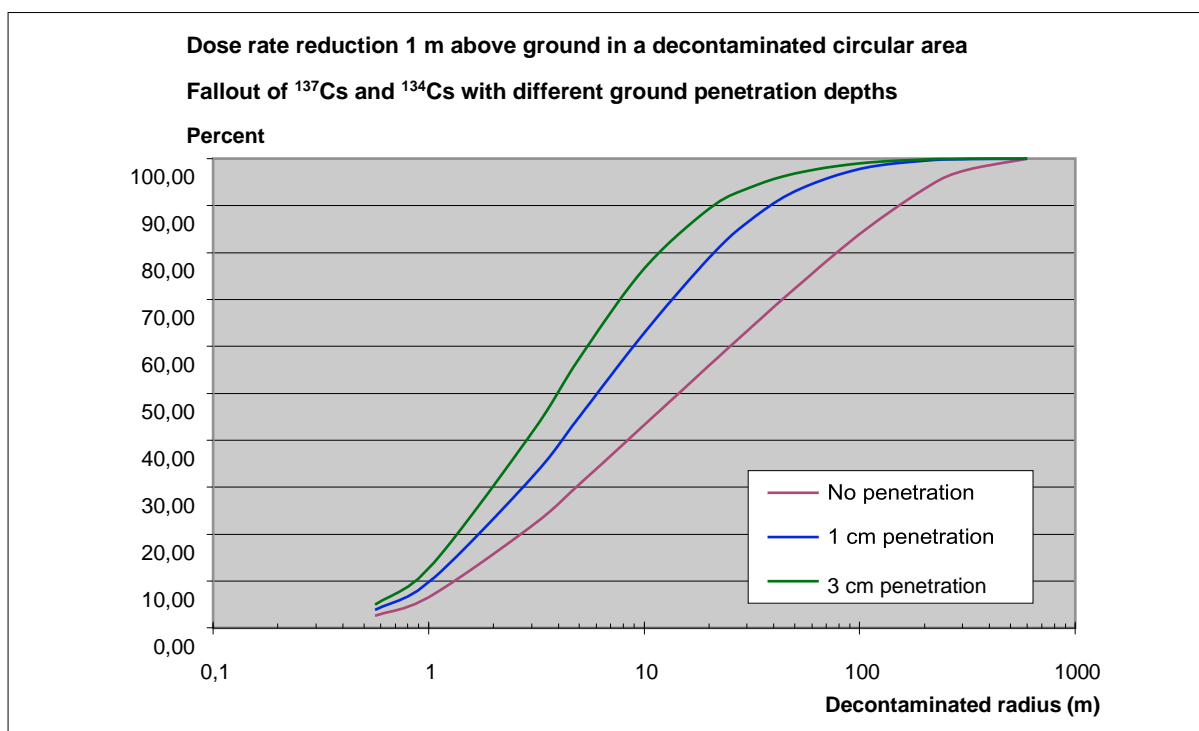


Figure A1. Relative decrease in air dose rate 1 meter above ground in the center of circular area from which all radioactive material has been removed. The curves show the decontamination efficiency dependence on the decontaminated radius as well as different penetration depths. The fallout is assumed to consist of equal amounts of activity from ^{134}Cs and ^{137}Cs . The model further assumes ground-/soil material with a density of 1600 kg/m^3 , attenuation coefficients for aluminum and build-up factors for air. An example of application: for a completely decontaminated area of 10 meters in radius in a large and plain field subjected to nuclear fallout the air dose rate in the center of circle will decrease by 43% if the fallout is situated on the surface, 65 % decrement if the radionuclides are distributed within the first centimeter of the ground and a 77% decrement if the radionuclides are distributed within the first three centimeters of the ground.

Physical structures, like buildings, can efficiently aid in decreasing the air dose rate by shielding. The shielding effect of buildings can be very complex since it dependent on geometry, physical properties of the building material and if the materials contain radionuclides. Even a minor contamination of radionuclides in building materials can lower the effect of the outdoor decontamination effect in indoor dose rate reduction.

Penetration of radionuclides into the ground will affect the dependence on the dose rate with varying

measurement altitude. The dose rate in a decontaminated area will be the lowest at minimum altitude. The dose rate will then increase with increasing altitude since radiation from the surrounding, contaminated, area have a straighter angle to the measuring point, i.e. does not need to pass large amounts of ground- and soil material and thereby avoiding significant attenuation effects. This effect is important to consider when decontaminating areas adjacent to tall buildings. It might be necessary to decontaminate a larger area to reach a sufficient dose rate reduction. If the altitude is increased further the dose rate will eventually decrease due to the increased attenuation of air. For a decontaminated circular area with a radius of 30 meters a maximum dose rate is expected on a 10-20 meter altitude if the ^{137}Cs is exponentially distributed in the ground with a mass depth distribution coefficient of 6.25 mm (Finck, 1992).

An important issue is how large area that needs decontamination in order to achieve a certain reduction in dose rate during a certain period of time. Figure A2 shows that the effect of decontamination efforts in the center of a decontaminated area is decreasing for every square meter that is decontaminated in the periphery. Assuming that the decontamination cost per square meter is independent of the distance, the decontamination efforts become less cost-effective for distances where people spend less time. It is more efficient to start a decontamination project adjacent to a new occupancy location than to expand a decontamination area.

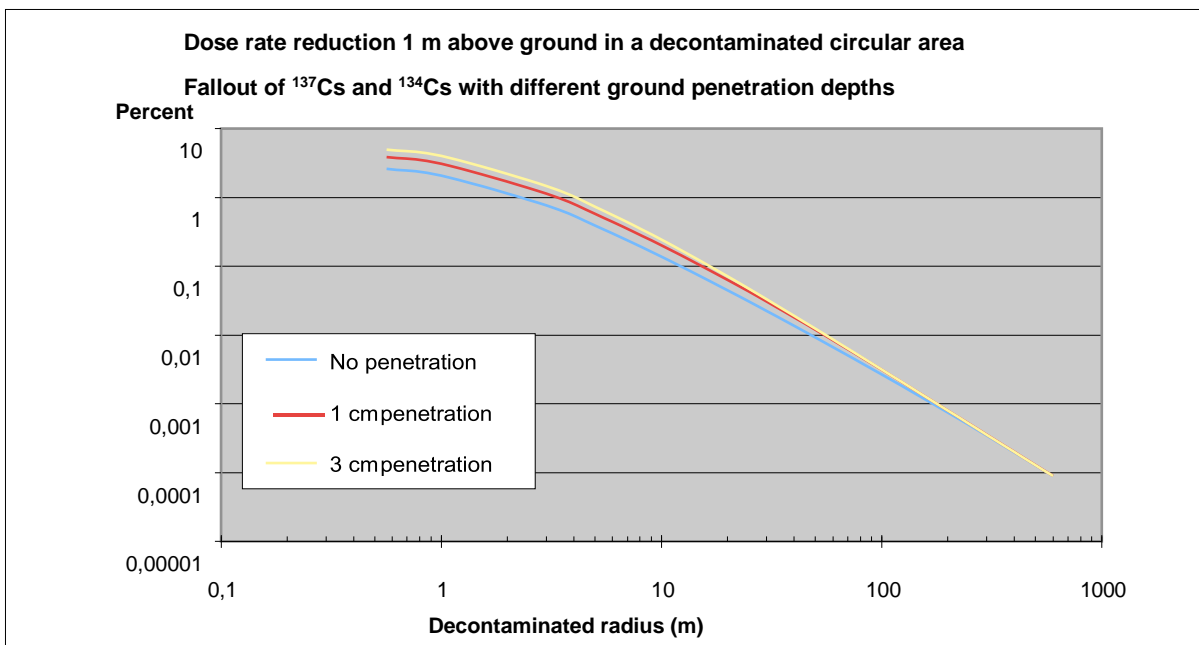


Figure A2. Relative reduction of the air dose rate 1 meter above ground per decontaminated square meter in the center of a circular surface from which all radioactive material of radiocaesium has been removed. The curves show the reduction of air dose rate as a function of decontaminated radius and three penetration scenarios. The fallout is assumed to consists of equal amounts of activity from ^{134}Cs and ^{137}Cs .

The area subjected to decontamination in order to achieve a certain dose reduction is thus dependent on multiple factors. Detailed measurements of the area radionuclide distribution are crucial before any decontamination can start. With the information obtained from the measurements, shielding factors of building and different efficiency of various decontamination methods it should be possible to design a model for choosing the optimal decontamination strategy for a contaminated area. Parameters further added into such model could include waste management and training of personnel.