Decontamination efficiency and waste generation for the decontamination of radioactively contaminated urban and rural environments

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Abstract

A radioactive fallout following a nuclear accident can result in contamination of large areas of land. In order to protect human health against ionizing radiation, large-scale decontamination that includes multiple sets of clean-up measures may be necessary. Sweden lacks national experience of this type of large-scale decontamination. There are thus great uncertainties in the effect of such a decontamination which is dependent on the efficiency and waste generation of the individual remediation measures. In this report, the results from a literature review of the Japanese experience of decontamination after the nuclear accident in Fukushima-Daiichi, 2011, are highlighted. We show that the Japanese decontamination efficiency was on average about 12 percentage points lower than the decontamination efficiencies listed in reference literature on radioactive material decontamination. Removed contaminated soil is by far the largest contribution to radioactive waste production during decontamination. There is a positive correlation between reduced radiation dose rate and amount of soil removed during decontamination. Over time, however, ecological processes contribute by far the most to reduced radiation dose rates. The results can be an important contribution to current decontamination strategies and valuable for responsible agencies and authorities in the case of nuclear fallout incident.

INTRODUCTION

A nuclear accident can result in airborne releases of radioactive material. It can give rise to ground depostion of radioactive substances. Several of the radioactive substances that can be released have short half-lives and decay so quickly (days or weeks) that no special decontamination is necessary. If, on the other hand, the more long-lived radionuclides ^{134}Cs (half-life 2.06 years) and ^{137}Cs (half-life 30 years) are released from the reactor, it would have a significant impact on the environment. ^{134}Cs and ^{137}Cs emit gamma radiation that reaches several hundred meters in air. These properties mean that the external dose rate from the ground deposition in a certain property in a neighborhood will to a large extent emanate from deposition in the neighboring areas, as the gamma rays can penetrate buildings and vegetation, and thus cause radiation exposure over many years to people who stay and live in the area. In a typical Northern European suburban neighborhood that has generally been contaminated with a ground deposition of ^{137}Cs, as much as 50% of the external dose rate inside a detached wood or brick building emanates from contaminated surfaces outside the own property (Hinrichsen et al., 2020). Through decontamination, e.g. scraping away the topmost (approx. 5 cm) ^{137}Cs contaminated soil layer, up to 90% of the radioactive contamination can be removed (Andersson, 2009). Decontamination of radionuclides is a costly process that, in addition to requiring large resources, also generates large amounts of waste (Munro, 2013; Hinrichsen et al., 2021). If a decontamination is to be carried out, it is therefore of great importance that the decontamination can be optimized. The optimization can take place by taking into account several different aspects, e.g. that it must be resource efficient in relation to how much damage to people and society can be avoided, while the amount of waste should be minimized.

Large-scale decontamination after a radioactive fallout has never been carried out in Sweden. There is thus a lack of knowledge and experience about what an authentic decontamination on a national level will entail in terms of resource consumption, decontamination efficiency and waste generation. In order to hypothetically estimate the outcome of a large-scale decontamination on Swedish terrestrial areas in terms of decontamination efficiency and waste generation, one can instead study the previously established methods that could possibly be used and which were developed through experimental trials and experiences after the decontamination in Ukraine and Belarus after the Chernobyl accident in 1986. Many of these methods are collected in the so-called EURANOS (European approach to nuclear and radiological emergency management and rehabilitation strategies) manual from 2011 (Nisbet et al.

2011). In the EURANOS manual, the decontamination methods are listed together with estimated decontamination efficiency, time consumption, necessary equipment and manpower. As some of the methods are developed and tested as dedicated experimental trials, there is a risk that the published values of the decontamination efficiency may be overestimated compared to the performance of the methods during authentic conditions. During the experimental trials the investigators took the time to methodically and systematically carry out the decontamination with good time margins (Andersson, 2009?), but it may be questionable whether these conditions may still prevail during real large-scale decontamination operations. However, accumulated experience from completed large-scale decontamination projects can be found in the plethora of literature published since the nuclear accident in Fukushima-Daiichi in 2011 (e.g. MOE, 2018). This study aims to i.) compare decontamination efficiency and potential waste generation between data from the EURANOS handbook and the results from the Japanese decontamination and to ii.) through the Japanese literature on decontamination measures obtain a realistic description of how a waste problem would appear in Sweden in the event of a large-scale decontamination work.

METHOD

In 2018, the Japanese Ministry of Environment (MOE) published a translation of a large work (published in the original language in 2017) summarizing the Japanese experience of the decontamination work after Fukushima-Daichii. The work, "Decontamination Projects for Radioactive Contamination Discharged by Tokyo Electric Power Company Fukushima Daichi Nuclear Power Station Accident", is a 500-page report with extensive descriptions, interviews, cost estimates and calculations all relating to the decontamination work (hereinafter referred to as "MOE (2018)"). The lead author of this report has read the Japanese report with the aim of identifying the most common decontamination methods and to study actual decontamination efficiency and waste generation. The identified decontamination methods from the Japanese report have been compared with the listed methods from the EURANOS handbook.

RESULTS AND DISCUSSION

Decontamination efficiency

The EURANOS manual describes more than 40 decontamination methods that result in a reduction of the external dose for outdoor environments. In the report from MOE (2018) it is stated that approx. 30 decontamination methods were used for outdoor environments. It is also stated in MOE (2018) that in the initial decontamination phase they started off by using the EURANOS handbook and applied the listed decontamination methods from this work. However, over time they adapted and optimized the methods for Japanese conditions. This adaptation could e.g. relate to differences in building construction, architecture, agriculture, etc., conditions that required a change in the execution of the decontamination method. Figure 1 shows the decontamination efficiency of a particular method from the EURANOS handbook compared to that measured in the decontamination in Japan. The Japanese decontamination efficiency is shown as a relative reduction (in percentage) of the collimated external radiation dose rate from ^{137}Cs one meter from the surface emitting the radiation. The comparison shows that, on average, the EURANOS handbook overestimates decontamination efficiency by 12 percentage points, reinforcing the image that the EURANOS handbook's methods and results were produced under more favorable conditions than those prevailing during the large-scale decontamination in Japan.

One of the most important decontamination methods to reduce external exposure from green and open ground surfaces is the removal of the top layer of soil (usually at least 5 cm). It is also this decontamination method that undoubtedly generates most waste. The difference in decontamination efficiency is large between the value stated in the EURANOS manual (94%) and that presented in MOE (2018) (58%), see Figure 1. The time between the fallout and the implementation of decontamination is critical to decontamination efficiency (Yasutaka et al., 2013); the shorter the time, the higher the efficiency in terms of relative dose reduction, which was also theoretically demonstrated in e.g. Rääf et al., 2020. This is because the deposited radionuclides only had time to vertically migrate a limited depth into the soil or were transported away by weathering processes. One can therefore assume that the high decontamination efficiency presented in the EURANOS manual applies to fresh fallout. The Japanese decontamination of the fallout-affected areas started about 6-12 months after the nuclear accident, and the measured decontamination efficiency (58%) is therefore not unrealistic. Decontamination efficiency is defined according to equation 1:

$$Decontamination\ efficiency\ (\%) = \left(1 - \left(\frac{Activity\ post\ decontamination}{Activity\ prior\ decontamination}\right)\right) \cdot 100 \qquad (1)$$

Roed et al. (1998) reviewed decontamination practices carried out in the summer of 1989 in 93 villages in the Bryansk region of Russia that were severely affected by the Chernobyl accident. The top layer of soil had essentially been removed there and sand or gravel had been added as radiation shielding. The time between the 1986 Chernobyl accident and the decontamination in 1989 may have contributed to the measured mediocre decontamination efficiency of 10–30%. Roed et al. (1998) believed that other reasons may also have contributed to the low decontamination efficiency, such as:

- Individual decontamination objects were not large enough in area (irradiation contributes from longer distances).
- Inadequate identification and method for decontamination of "hot-spots" such as e.g. the ground below gutters and surrounding ground around buildings.
- Insufficient amounts of topsoil were removed.
- Insufficient accuracy in the decontamination work.

Considering the relatively low decontamination efficiency observed by Roed in the Bryansk area, a decontamination experiment was carried out in Sweden in connection with the nuclear accident exercise Demoex 2006 in Halland. The intention was to test the best possible technology available in Sweden. The short-lived radioactive substance $^{99}Tc^{m}$ (half-life 6 hours) was used as a radioactive pollutant, which was spread over a building and surrounding land. Personnel with special training in decontamination handled the machines, such as wet padding. Around the building, the soil material was removed down to 10 cm with an excavator. Parts of the work were carried out by hand to avoid spillage of radioactive material. The decontamination efficiency was measured at 90 - 98%. The high efficiency was mainly due to extreme care in the work to prevent spillage and mixing of materials between contaminated and clean areas. The experiment showed how important it is to have well-trained personnel for the methods used and that all work steps are done in the correct order (Finck et al. 2008).

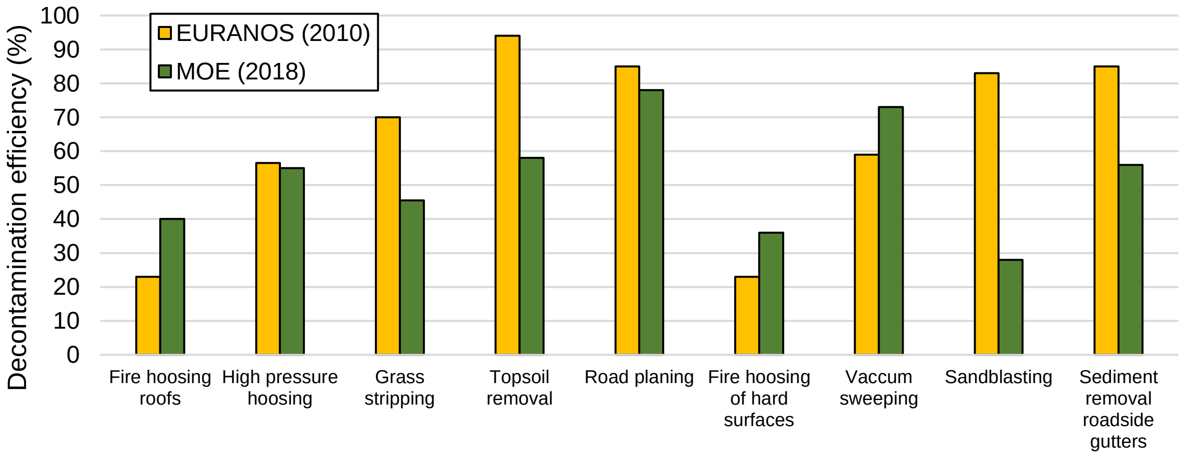

Figure 1. Comparison of the mean decontamination efficiency given in the EURANOS manual (2010) and measured values in Japan after the Fukushima-Daichii nuclear accident (MOE, 2018).

According to measurement results obtained on 30th of June 2017 after decontamination in the so-called Special Decontamination Area in Japan decontamination efficiencies of 73% for urban areas, 46% for forest land, 68% for agricultural land and 61% for roads and pavements could be estimated. It was observed that the higher the radiation dose rate before decontamination began, the greater decontamination efficiency was achieved. It could also be seen that the greater the amount of soil that was removed around a studied point, the greater was the decontamination efficiency (MOE, 2018).

Figure 2 shows the predicted volume amount of waste per June 2013 divided into different classes and activity concentrations. According to recent publications, this waste estimate matches well with the real outcome (Evrard et al. 2019). It appears that decontamination waste in the form of soil masses accounts for a predominant proportion of all waste (93%). Ash is derived from burning contaminated combustible waste, mostly plant material from gardens, parks, forests and farmland. Other waste accounts for a small percentage of the total amount of waste (<1%) and consists of construction materials such as e.g. roofs, walls, building materials, etc. Soil with a lower activity concentration (<8000 Bq/kg) is planned to be reused as filling material in infrastructure projects (Evrard et al. 2019). However, the Japanese authorities have decided that only filling material with an activity concentration below 6000 Bq/kg may be used if it is buried under at least 50 cm of clean soil. This means that approx. 10 million cubic meters of soil in the lower activity class will become usable as filling material in approx. 2024. Takai et al. (2018) suggest that a reclassification of the highly active (>8000 Bq/kg) soil masses may allow additional amounts of soil to be reused in the long run. All other waste must be stored in specially designed facilities pending the decay of ^{137}Cs activity.

To minimize amounts of waste (mostly soil) and its related costs, Yasutaka et al. (2013) suggest either deep plowing or three-step digging, where in both methods the contaminated soil is turned deeper into the soil layer. However, this method is considered more suitable for agricultural land and less useful in urban areas.

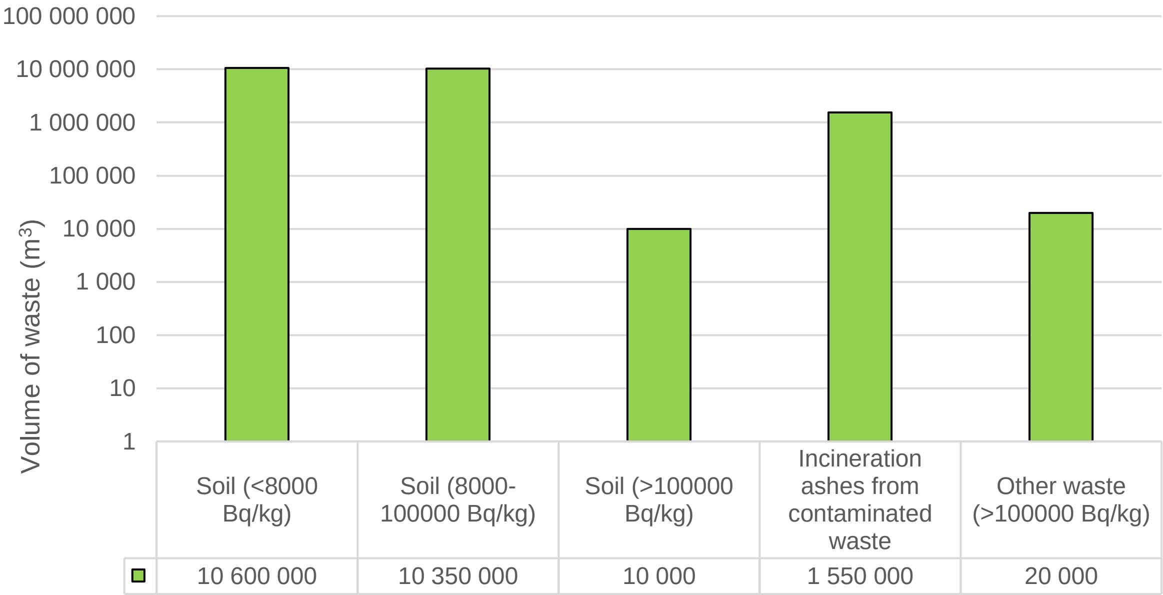

Figure 2. Estimated amount of waste (July 2013) from the decontamination after Fukushima, Japan. Volume-reducing measures are also included here, such as e.g. combustion. The figure is reproduced and modified from MOE (Fig. 3-21. 2018).

Figures 3A and 3B show how the radiation dose rate decreases in relation to the amount of soil removed as a result of decontamination measures in ten municipalities in Japan. The figures show that larger amounts of soil per surface unit need to be removed to reach a radiation dose rate below 1 µSv/h for the most contaminated municipalities (points A and B, Figure 3A). By making a linear regression of the plot in Figure 3B it can be shown that the radiation dose rate was reduced by 1 µSv/h for every 16 m^3 per 1000 m^2 of soil removed. This can be translated into the radiation dose rate being reduced by 1 µSv/h for every 1.6 centimeters dug away (16 m^3/1000 m^2 = 0.016 m). From Figure 3 it can be estimated that the decontamination workers never dug deeper (on average) than just over 6 cm (point A), while they always dug at least 0.5 cm (point J). All in all, for all ten municipalities, they dug on average 3.7 cm deep when decontaminating.

It should be noted that it is technically very difficult to mechanically remove thin layers of soil material without disperse parts of the radioactively contaminated material onto the clean surface. It is also difficult to remove all radioactive material around plants. This, together with the fact that ^{137}C is subsequently carried further down into the ground through natural processes such as infiltration with rainwater, chemical processes and biological life (ecological processes), may explain why it is necessary to dig deeper to achieve higher decontamination efficiency, especially if the decontamination work begins several years after the fallout.

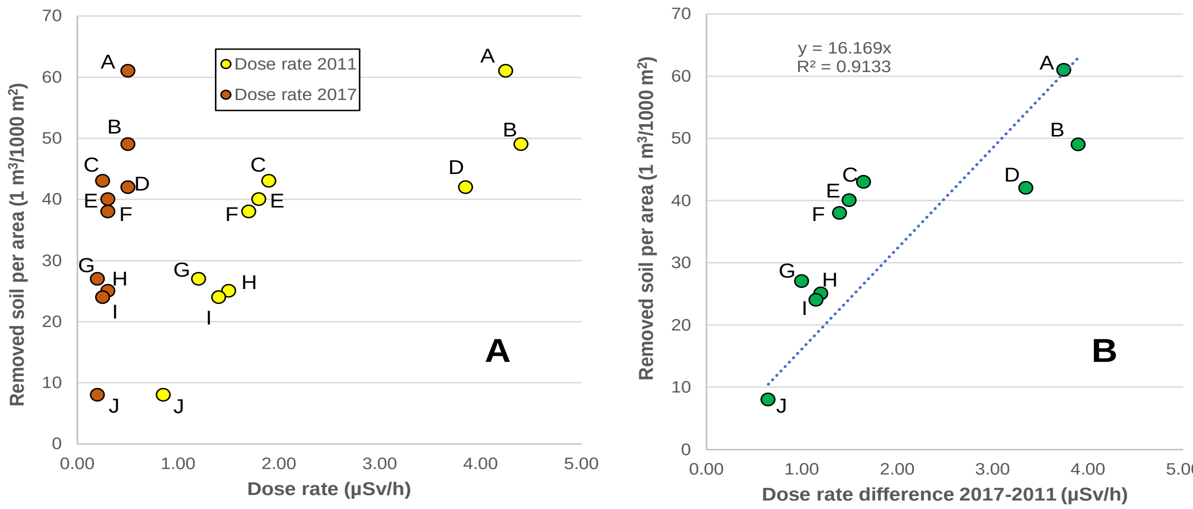

Figure 3. Difference in radiation dose rate versus removed volume of soil per unit area. **A** shows the measured average radiation dose rate in 10 municipalities before (yellow markings) decontamination (2011) and after (red markings) decontamination (2017) versus generated amount of soil waste per unit area. **B** shows the difference in radiation dose rate between 2011 and 2017 against the generated amount of soil waste per unit area. Each point in **A** and **B** corresponds to a Japanese municipality where decontamination took place. The figures are reproduced and modified from MOE (Fig. 5-24. 2018).

When describing the effectiveness of decontamination efforts, one should also take into account that the external radiation dose contribution from ^{134}Cs and ^{137}Cs decreases over time, not only through the physical decay of the isotopes, but also because ecological processes such as deep migration and erosion lead to a further decrease in the radiation dose contribution above ground. According to what emerges through: 1) our own model calculations (Rääf et al. 2020), 2) initial Japanese measurements and modeling of the external dose contributions from ^{134}Cs and ^{137}Cs made by Kinase et al. (2014), as well as 3) Japanese measurement series with car-borne radiometry between the years 2011 and 2017 published by Andoh et al. (2020), the external dose rate in the fallout-affected areas outside Fukushima would decrease by about 85% between the years 2011 and 2017 without any applied decontamination measures. Given this reduction (which had thus occurred regardless of the measures), it can be estimated that the reductions in dose rate for sites A to J as reported in the MOE (Fig. 5-24. 2018) were only achieved to about 5% through the decontamination efforts.

CONCLUSIONS

In general the decontamination efforts after the Fukushima-Daiichi accident show a lower decontamination efficiency than those listed in the reference literature on decontamination of radioactive materials (eg EURANOS handbook) and that achieved in a limited Swedish experiment (Finck et al., 2008). This needs to be considered when planning for a possible decontamination in Sweden in case of a future large-scale deposition of gamma emitting radionuclides such as ^{137}Cs. It can be assumed that the Japanese decontamination efficiency is more representative of what can be achieved in large-scale efforts involving more than 1000 km^2 urban and rural land. The experience from Japan consists of many observations per decontamination method and is based on authentic operating conditions with all of its constituent uncertainty-affecting variables such as time pressure, varying skills of decontamination workers and limited resources of various kinds. The largest waste category consists of soil masses (93%) removed during the decontamination. This waste category is also one of the more resistant to volume reduction measures (compared to combustible waste, for example), which means that waste volumes will only be able to be reduced marginally. Good planning for the storage of large amounts of soil is required in the event of a possible decontamination on Swedish land, and suggested plans for interim storage and repository are under preparation (MSB, 2022).

The differences in decontamination efficiency between results from controlled experiments and the actual results obtained in the former The Soviet Union after the Chernobyl accident and in Japan after the Fukushima accident show how extremely important it is that all decontamination personnel are trained for their tasks, that the work is carried out carefully according to proven methods and that the decontamination is carried out without delay if the intention is for people to continue to live and work in the area.

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