A Systems Engineering Analysis

RADIOACTIVE SOIL HANDLING AND REMEDIATION PROCESSES AT FUKUSHIMA DAIICHI

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Contents

Co	onter	nts	i
\mathbf{Li}	st of	Figures	ii
\mathbf{Li}	st of	Tables	ii
1	Intr	roduction	1
	1.1	Fukushima Daiichi Accident	1
	1.2	Aftermath in Fukushima Prefecture	1
	1.3	Radiation Hazards and Risks	1
		1.3.1 Engineering Controls	2
		1.3.2 Soil Risk Management	3
2	Rad	diation Levels	3
	2.1	Monitoring Data	3
	2.2	Methodology and Comparisons	3
		2.2.1 Cultural Considerations	4
		2.2.2 Comparison through Quality Control	4
		2.2.3 Statistical Comparison	4
	2.3	Radioactive Decay	6
		2.3.1 Clean-Up Event Correction	6
		2.3.2 Predictive Modelling	6
3	Site	e Characterisation	7
	3.1	Radiation Mapping	7
	3.2	Soil and Radiation Distribution	8
	3.3	Calculating Soil Removal	9
		3.3.1 Impact Factor	11
		3.3.2 Limitations at High Doses	11
		3.3.3 Removal Volume	12
4	Mat	terial and Energy Impact	12
	4.1	Removal Process	12
		4.1.1 Raw Material Consumption	12
		4.1.2 Energy and Mass Flow of Process	13
5	Out	tcomes, Cost and Alternatives	15
	5.1	Life-Cycle Cost	15
	5.2	Alternative Actions	15
	5.3	Conclusion	15
6	Ref	ferences	16

List of Figures

2.2.1 Bland-Altman quality control data comparison	5
2.3.1 Station 6 radiation dose modelling and correction	6
2.3.2 MP4-MP5 radiation dose and predicitve mdoelling	7
3.1.1 Grid applied to Fukushima precinct	3
3.1.2 3D surface illustrating radiation dose across Fukushima	3
3.2.1 Percentage contributions of radioactive decaying particles	9
3.3.1 Time to radioactive decay with and without removal)
3.3.2 3D object plot demonstrating volume of soil to be removed by location	2
4.1.1 Energy-mass flow of removal and storage system	4

List of Tables

1.3.1 Hazard and risk identification of radiation at Fukushima	2
2.1.1 Monitoring Station Example Measurements	3
2.2.1 Summary of Wilcoxan Signed Rank Test for June 2011	õ
2.2.2 Summary of Wilcoxan Signed Rank Test for July 2011	5
2.2.3 Summary of Wilcoxan Signed Rank Test for August 2011	õ
2.3.1 Summary of monitoring post predictive models	7
3.3.1 Earliest dates to acheive safe radiation levels	1
3.3.2 Summary of currently required top-soil removal depths	1
4.1.1 Material audit and costing of removal and storage 14	4
5.1.1 Overall life-cycle cost of removal, storage and decommissioning	5

Abstract:

As a result of the Fukushima Daiichi nuclear accident in 2011, surrounding soil remains contaminated with harmful radioactive isotopes. Handling this soil is a major engineering challenge in the clean-up operation and this paper provides an analysis of soil remediation in the immediate areas. Determining the volume of soil to be handled, and the material, energy and storage requirements formed the research question. Based on historic radiation dose measurements throughout the Fukushima site, a series of statistical and analytical methods were used to develop a radiation model. The results of ongoing research allowed top-soil removal, required to mitigate radiation dose, to be computed. Consideration of material and energy consumption led to recommendations for improved processing and consideration of alternate methods. Ultimately the efficiency of a simple removal and storage system was questioned and the combination of multiple methods proposed as necessary in search of an effective clean-up operation. Analysis of the entire soil handling system allowed total cost and soil removal to be calculated based on current decay rates, thus aiding in the planning process of the Fukushima clean-up operation.

Recommendations:

- **R1** Engineering controls and design should be maximised when planning for the high priority elimination of radioactive soil hazards at Fukushima Daiichi.
- R2 Tokyo Electric Company's (TEPCO) ongoing recordings or radiation dose throughout the Fukushima precinct are reliable and extensive measurements suitable for modelling and predictive purposes.
- **R3** The exponential distribution of radionuclides favouring the near-surface region means top-soil removal is the most viable and efficient elimination remediation method at Fukushima Daiichi.
- **R4** The decommission operation must include the preparation of storage sites within a 15km radius, capable of storing 778710m³ of waste, at minimum until 2039.
- **R5** Investigate reprocessing nuclear material to concentrate radioactivity in a smaller volume, thus reducing raw material consumption and embodied energy of the removal process.
- **R6** Funds should be prepared for an 82 billion JPY operation to remove soil in the immediate Fukushima precinct, remediate environmental damage, reduce health risks, and restore land for farming purposes.

1 Introduction

The release of radioactive material during and following the Fukushima Daiichi Nuclear Power Plant accident in Japan has demanded an immense clean-up operation. The surrounding land and ocean area has been contaminated with significant levels of radioactive elements and could take decades to restore to reasonable levels (Chen 2011). In particular, the large land mass over which radiation spread in the immediate aftermath of the accident means a large volume of previously farmed or industrial soil must be carefully handled in the coming years. The cost of this operation is vast but secondary to safety and the enormous human and material resources consumed in soil recovery. Understanding the systems in place to handle radioactive soil, their effectiveness and efficiency, is critical to determining the long term impacts on human life in the region and evaluating the true impact of the Fukushima accident.

1.1 Fukushima Daiichi Accident

The March 2011, magnitude 9.0 earthquake and subsequent tsunami crippled the Fukushima Daiichi Nuclear Power Plant, located on Japan's east coast (Kim 2014). Following the loss of external power to the plant due to the earthquake destroying power lines, a tidal wave inundated the basement buildings which housed the back-up diesel generators and sea-water pumps used for cooling the reactors (Omoto 2013). Consequently, with no active cooling the temperature inside the 3 reactors operational at Fukushima at the time, rose and the zirconium fuel cladding began mixing with water vapour, producing hydrogen gas within the reactor (Hoffman 2012, Omoto 2013). When the pressure inside Unit 1 became too great, an explosion occurred, destroying the top of the reactor building and exposing the nuclear core (Song 2013). Unit 3 suffered a similar fate shortly after, whilst Unit 2 observed a sudden pressure drop which has since been attributed to a rapid release of radioactive material to the environment (Kim 2014, Omoto 2013). Unit 4 which was not active at the time, but contained recently spent nuclear fuel in cooling ponds, also suffered damage and threatened to release large volumes of nuclear material into the atmosphere (Song 2013). The combined explosions and continued ejection of radioactive material from the plant had severe effects not only on the environment but also the surrounding townships which were evacuated and have not since returned (Evrard 2015).

1.2 Aftermath in Fukushima Prefecture

Nuclear fission generates electricity by forcing a large element's nucleus, typically uranium, to split into smaller fragments whilst releasing heat energy (Chen 2011). Whilst safe in a controlled environment, once exposed like at Fukushima Daiichi, the fission products can be harmful to human health, particularly caesium-134 and caesium-137 which hold long radioactive half-lives (Brookins 1984). Alarmingly, nuclear material continues to leak through the damaged floor of the reactor buildings, and the initial explosion which deposited Cs-134 and Cs-137 into the air, surrounding soil and ocean, also remains an issue today (Hardie 2014, Song 2013). The soil which surrounds the Fukushima site comprises varying levels of radioactivity and its handling is a key part of the clean-up and restoration of the site. The radiation emitted during the decay of radioactive isotopes such as caesium is particularly damaging to human health and is known to cause mutations in cells at high enough doses (Harada 2014). Consequently, displaced villagers and farmers cannot return to the area without effective soil handling and workers part of the decommissioning of the plant are exposed to further radiation until the soil is removed. Thus, the purpose of this paper is determining the *magnitude, nature* and *impact* of this clean-up process, providing planning recommendations based on modelling and systems engineering-based analysis.

1.3 Radiation Hazards and Risks

Fukushima in its current state poses a series of radiation hazards, all of which stem from the nuclear material which was designated for powering the reactors during their life-time. Whilst accessible fuel was quickly removed from the site or placed in cooling ponds in the immediate aftermath of the accident (Baba 2013, Pint et.al. 2013), the severe damage inflicted by the March 2011 earthquake, tsunami and subsequent explosions at the plant mean the most concentrated radioactive isotopes continue to leak from the reactor buildings. Water which was pumped into the reactors in response to the nuclear meltdown is known to have leaked into the water table and ultimately the ocean (Tsumune et. al. 2012). Similarly, the damaged floor of multiple reactor units means the surrounding water supply and soil could be contaminated with radioactive nuclides for decades to come. These form secondary sources of radiation and are further hazards (see Table 1.3.1) to both the general Fukushima population and the workers completing the decommissioning operation.

Hazard	Risk	Likelihood	Severity	Level	Control	Further Comments
Spent fuel	Exposure	Unlikely	Significant	Medium	Isolation	Held in cooling ponds until signifi- cantly decayed
Unused fuel	Exposure	V Unlikely	Severe	Medium	Elimination	Unused fuel is removed from the Fukushima site for use at other fa- cilities
Radioactive wa- ter	Ingestion	V Unlikely	Severe	Medium	Admin	Water sources in the region are signposted not for drinking
	Agriculture	Unlikely	Significant	Medium	Isolation, Engn.	Avoid leakage of cooling water etc. into water table
	Supply	V Unlikely	Significant	Medium	Isolation, Engn.	Prevent contaminated water enter- ing steams etc. which feed water community supplies
	Ecosystems	Likely	Moderate	Medium	Isolation	Ice barrier to sea under construc- tion to prevent flow of water into ocean
Radioactive soil	Exposure	Possible	Significant	Med-High	PPE, Engn.	Workers handling soil on-site wear full-body hazard suits
	Agriculture	Possible	Significant	Med-High	Elim., Engn.	Remove top-soil from affected ar- eas
	Dispersion	Possible	Significant	Med-High	Elim., Engn.	Radioactive soil becomes mixed with clean soil, diluting radiation but increasing affected area
Atmospheric ra- diation	Exposure	Admin/PPE	Likely	Minor	Medium	Highest does areas are restricted, lower dose areas are patrolled but effects are limited at these doses
Waste storage	Leak	Possible	Severe	Med-High	Engineering	Concentrated waste in tanks or stockpiles is carefully monitored by sensors

Table 1.3.1. Hazard identification and associated risks in the Fukushima clean-up mapped to the hierarchy of controls

1.3.1 Engineering Controls

The greatest risk posed by radioactive material such as spent fuel, and contaminated soil is prolonged exposure to humans at high doses. Low levels of radiation are present in everyday life as a result of cosmic background radiation however the Fukushima precinct, much like Chernobyl in Ukraine exhibits far higher doses of radiation (Steinhauser et. al. 2014). Researchers have also reported that contaminated soil and water in agricultural areas has resulted in the production of food which is inherently radioactive (Unno et. al. 2014). Meanwhile, workers on the Fukushima site are fitted with hazard suits as a form of personal protective equipment (as noted in Table 1.3.1), however the Japanese government recognises this lies at the bottom of the risk control hierarchy. By removing contaminated soil, and employing the more preferable engineering and elimination control methods it is hoped the risk of radiation exposure can be greatly reduced and the Fukushima site restored. **R1** Engineering controls and design should be maximised when planning for the high priority elimination of radioactive soil hazards at Fukushima Daiichi.

1.3.2 Soil Risk Management

The greatest challenge is the magnitude of this operation, Japan has already declared a 20km radius which many believe will never be properly restored, and some research suggests a land radius in excess of 80km could be unsuitable for farming (Nadesan 2012). This equates to an immense volume of soil which will require storage for decades along with debris and nuclear waste from the Fukushima plant itself. Thus, this investigation focuses on determining which zones within the immediate area should employ top soil removal and to what degree, based on both cost and material impact factors. Radioactive isotopes do progressively decay as they emit radiation, opening the possibility to simply abandoning areas and waiting for levels to decay (Brookins 1984). Thus, quantitatively balancing this with the need to restore areas and ensure safe working conditions substantiates the following investigation.

2 Radiation Levels

2.1 Monitoring Data

The fraught nature of nuclear accidents and the ability of radiation to ensue widespread harm means monitoring stations are well established at all nuclear power plants, including Fukushima Daiichi. Tokyo Electric Power Company (TEPCO) which operates the Fukushima plant (and is now subsidised by the Japanese government) compiles monitoring data throughout the precinct which is readily available in both csv. and map format. For the purposes of developing a soil re-mediation model, data was gathered based on radiation levels at specific locations on a monthly basis. TEPCO does report radiation dosage throughout the plant on an hourly basis but values are relatively constant and the interest here is on the long term impacts. In the immediate aftermath of the accident a total of 17 stations were operable across 15 locations. 8 monitoring posts (MP1-MP8) operated from March 2011 through to October 2012, whilst 7 permanent stations record data indefinitely (up to September 2016). The full set of monthly data considered in this study has been organised and tabulated, and can be viewed on request from the Appendix; example measurements demonstrating data organisation are shown in Table 2.1.1.

Table 2.1.1. Radiation dose (μ Sv/h) at Stations 1-7 and Monitoring Stations 1-8 on 01/10/2012

S(2)	S(3)		S(7)		S(4)	S(6)	S(1)	S(5)	MP1	MP2	MP3	MP4	MP5	MP6	MP7	MP8
371.9	7.2	7.0	28.3	20.0	114.0	205.0	2015.0	427.0	3.5	6.1	7.3	6.9	6.9	3.8	7.7	5.8

2.2 Methodology and Comparisons

The measurement of radiation can vary significantly between sources, and is often specified using different quantities. The most common measures are activity, measured in Becquerel (Bq), which describes the amount of radioactivity in a sample, and Sievert (Sv) which measures the dosage delivered from a sample (Knoll 2010). Since the risk of radiation to humans and livestock, through contaminated soil, is the focus of this analysis, the later measurement is preferred here. TEPCO who ran Fukushima during its operational lifetime are also responsible for its decommissioning, and publish radiation levels throughout the site in μ Sv/h, or the effective radiation dose per hour. Despite the detailed and extensive measurements taken by TEPCO significant suspicion has arisen about the accuracy of these measurements (Nadesan 2014), particularly in the immediate aftermath of the accident.

2.2.1 Cultural Considerations

Scholars have suggested that TEPCO and the Japanese government did attempt to disguise the magnitude of the disaster both to reduce panic in evacuated residents and foreign nations (Shrader-Frechette 2012, Green 2012). Particular concerns arose because despite an established and respected position, Japan has a history of bureaucracy and misleading behaviour in disaster scenarios. Social researchers have noted Japan for being a tightly knit community which experiences a high level of collectivism but also a strong sense of hierarchy in government and large corporations (Shrader-Frechette 2012). Several large cover-ups such as defects in Mitsubishi vehicles in 2000 (Pejovic 2010), have been exposed in the past and TEPCO later admitted that the vastness of the meltdown was not immediately realised (Green 2012). There is also precedence for accidents which have been triggered by an over reliance on tradition norms, hierarchy and respect for superiors. The Amagasaki rail accident was partially attributed to pressured workers being in fear of their superiors, company punishment and not feeling competent with reporting procedures (ARAIC 2007). This all calls for caution when considering the data provided by TEPCO, however in the months and years after the accident, significant efforts have been made to increase transparency.

Whilst operating as an energy supplier, TEPCO was a profit driven organisation, however the Fukushima accident effectively ended this business operation. The enormity of the clean-up and immense clean-up will far exhaust TEPCOs funds or responsibility, yet as the owner and operator of the plants, they comprise the most technical knowledge applicable to the decommissioning process (NEA2012). Thus, TEPCO is now supported by government funding and in essence Japan has declared all funds necessary for the clean-up operation will be provided through a subsidiary (the Nuclear Damage Compensation and Decommissioning Facilitation Corporation) (NDF2016). This largely eliminates any need for secrecy, TEPCO no longer comprises any financial gain or brand image connections with the Fukushima accident and has negligible motivation for providing misleading radiation measurements. Oversight from both the Japanese Atomic Energy Agency and the International Atomic Energy Agency also ensures the accuracy of radiation data. However, to further validate the extensive TEPCO data, a subset was compared to a sample (although far smaller in size) measured by the Ministry of Education, Culture, Sports, Science and Technology (MECSST), Japan.

2.2.2 Comparison through Quality Control

Time-series graphs are typically used in process engineering to highlight when a system or activity is completed successfully, within a tolerance. This type of quality control analysis is applied here to consider the validity of the TEPCO derived data. By comparing 3 subsets of the TEPCO data with a third party independent source in a scatter chart, bias can be interpreted qualitatively. Data from Stations 3 and 7, along with MP1-MP6 was compared with values published by MECSST. The set of stations compared was limited by the ability to match up MECSST measurements (which were described in terms of kilometres from the Fukushima reactors) to the location of the TEPCO stations (see Tables 2.2.1-2.2.3). Values at the 9 comparable locations were considered for 3 successive months shortly after the accident (June, July, and August 2011) and the mean and difference of each pair was computed (using descriptive statistics methods) and plotted on a series of Bland-Altman graphs (see Figure 2.2.1). A clustering significantly above or below 0 is a typical visual indicator of bias, which was not present in any of the selections. Upper and lower limits were also set as 2 standard deviations from the mean (95% confidence) on the basis of confidence intervals. Across all 3 dates, only 2 pairs of measurements neared this cut-off, and thus the TEPCO data was considered representative of the true values.

2.2.3 Statistical Comparison

To confirm the accuracy of the extensive TEPCO data set, a selection of measurements were compared via statistical means. Data sets which are 'normal' are typically compared through t-tests and student's t-test. However, a correlated-sample t-test assumes that the difference between paired values is random, the source population is approximately normal, and the measurement scales of the two methods are split into equal intervals. As the radiation measurements are location based they cannot be assumed normal, and given that TEPCO makes recordings at ground stations but MECSST made airborne measurements with correction factors, a t-test is not suitable.



Figure 2.2.1. Bland-Altman plots of mean and difference between doses at Stations 3 & 7, MP1-6, and MECSST aerial readings on three dates in the aftermath of the accident, demonstrating the quality control comparison conducted

Furthermore, since the measurements at a selection of stations on 3 dates are of interest, the pairs cannot be said to have been arbitrarily picked. However, the Wilcoxon Signed-Rank Test can perform the same type of equivalence test on matched pairs whilst remaining non-parametric. In order to perform such a test, the data from the 3 different dates is considered independently but with the same null and alternate hypothesis:

- **H**₀: TEPCO and MECSST measurements share equivalent population mean ranks and thus are likely derived from the same population
- **H**_A: TEPCO and MECSST measurements comprise different population mean ranks and thus cannot be assured to be from the same population

A Wilcoxon Signed Rank Test was performed for the same three sets of comparative data seen in Section 2.2.2, evaluating the absolute difference of each pair, ranking the differences, reassigning the direction of the rank (positive or negative) and summing the result to form W (see Tables 2.2.1-2.2.3). For 95% confidence (p = 0.05) in a Wilcoxon Signed Rank Test comparing 9 matched pairs of data, a standard look-up table provides 29 as the point of statistical significance. For absolute values of W (|W|) less than 29, the null hypothesis cannot be rejected, thus suggesting that the two data sets could resemble equivalence. In all 3 cases this condition was satisfied, indicating that the TEPCO and MECSST data share equivalent population mean ranks. There was potential to perform additional statistical tests to further demonstrate equivalence, however this result combined with the previous qualitative analysis, instilled a high degree of confidence in the validity of TEPCO data. Since the more extensive TEPCO recorded data was shown as representative of third party measurements, it was reasonable to develop the analysis on the basis of these results.

Table 2.2.1. 01-06-11 vs. 03-06-11

	X_a	X_{b}	Diff.	S/R
S(3)	13.0	13.6	0.6	-5.5
S(7)	28.3	29.6	1.3	-7
	31.0	29.6	1.4	+8
MP1	5.0	4.8	0.2	+1.5
MP2	22.0	22.2	0.2	-1.5
MP3	14.0	14.6	0.6	-5.5
MP4	13.0	11.0	2.0	+9
MP5	16.0	16.3	0.3	-3
MP6	35.0	34.6	0.4	+4
		Total	= W	0
			$ \mathbf{W} =$	0 < 29

Table 2.2.2. 01-07-11 vs. 30-06-11

X_a	X_{b}	Diff.	S/R
13.8	14.6	0.8	-8
28.2	27.6	0.6	+5.5
35.0	36.6	1.6	_Ç
5.0	5.3	0.3	-2.5
24.0	24.7	0.7	-7
15.0	14.6	0.4	+4
14.0	14.6	0.6	-5.5
17.0	16.8	0.2	+1
37.0	36.7	0.3	+2.5
	Total	= W	19
	1	W = 1	9 < 29

Table 2.2.3. 01-08-11 vs. 05-08-11

X_a	X_b	Diff.	S/R
14.7	14.4	0.3	$^{+1}$
82.0	83.9	1.9	-7
42.0	41.2	0.8	+5
5.0	4.6	0.4	+2.5
24.0	24.5	0.5	-4
15.0	17.2	2.2	-8.5
15.0	14.6	0.4	+2.5
18.0	19.1	1.1	-6
39.0	41.2	2.2	-8.5
	Total	= W	23
	D	N∣ = 2	3 < 29

R2 Tokyo Electric Power Company's (TEPCO) ongoing recordings of radiation dose throughout the Fukushima precinct are reliable and extensive measurements, suitable for modelling and predictive purposes.

2.3 Radioactive Decay

In order to calculate soil removal, particularly over a decade long clean-up operation, the natural radioactive decay of isotopes must be considered. Theoretical relationships for radioactive decay based on the initial activity and half life of an isotope are well established, and form an exponential relationship (Brookins 1984). Whilst this ideal behaviour is not always observed in nature, radionuclides more closely emulate theoretical phenomena than many other physical concepts. Thus, when characterising the radiation levels at Fukushima overtime, a strong correlation with exponential decay was expected. The radiation measurements published by TEPCO were plotted against time, stretching from May 2011 through to September 2016 and an example of the results is shown in Figure 2.3.1 (*left*), which highlights the exponential reduction in radiation levels at Station 6.



Figure 2.3.1. Radiation dose measurements south of the administration building (Station 6), (*left*) demonstrating a clean-up event in 2015 which significantly reduced dosage and (*right*) the corrected rate of natural radioactive decay ignoring this event.

2.3.1 Clean-Up Event Correction

It was noted that at Station 6 and several other locations, radiation levels reduced as expected for the majority of the period, but a sudden drop was also present. This drop was traced to February 2016 in the case of Station 6 and is likely the result of TEPCO completing some re-mediation action. The removal of radioactive soil or nuclear material nearby Station 6 during this month is the most likely cause. Whilst this is important in terms of the overall radiation levels at several locations, it must be excluded from the natural rate of radioactive decay. Thus Figure 2.3.1 (*right*) shows a corrected curve whereby the sudden drop in radiation has been removed, further improving the model for radioactive decay to an \mathbb{R}^2 value of 0.98. Similar procedures were undertaken for MP7, MP8, and Station 7.

2.3.2 Predictive Modelling

Whilst Stations 1-7 have recorded data since the accident to present day, the Monitoring Posts (MP1-MP8) were switched off in October 2012. Since the 7 basic stations are not sufficient to form a soil removal model, predictions about the radiation levels at these MP sites since 2012 had to be made. The exponential regression that was applied can be seen in Figure 2.3.2, and the predicted values for all the MP sites for each month since October 2012 are included in the Appendix. A strong correlation was found in nearly all of the regression models, with 3 exhibiting coefficients of determination >0.9 and all bar MP1 exceeding 0.82. Considering the relatively small sample size of the MP stations, and the complexity of exponential regression, these are strong relationships. A summary of the final mathematical models employed in predicting radiation doses are detailed in Table 2.3.1. Combining these results with the extensive station data means the radiation levels throughout the Fukushima site can be considered for

the entire period March 2011 - September 2016, and into the future through predictive modelling. This allows the radiation levels which have to this point been considered in isolation to be mapped in accordance with their location.



Figure 2.3.2. Radiation dose measurements at Monitoring Posts 4 (*left*) and 5 (*right*), with an outlier (initial value) omitted from each in order to accurately predict long-term radioactive decay at MP4 and MP5.

	Adjustments	R^2	Regression Equation
MP1	none	0.7682	$y = (2.0729 \times 10^{17}) e^{(-9.3877 \times 10^{-4})x}$
$\mathbf{MP2}$	none	0.8944	$y = (1.0564 \times 10^{64}) e^{(-3.5381 \times 10^{-3})x}$
MP3	outlier removed	0.9324	$y = (3.5918 \times 10^{30}) e^{(-1.6601 \times 10^{-3})x}$
MP4	outlier removed	0.9345	$y = (1.4706 \times 10^{30}) e^{(-1.6397 \times 10^{-3})x}$
MP5	outlier removed	0.9267	$y = (1.6041 \times 10^{40}) e^{(-2.2337 \times 10^{-3})x}$
MP6	outlier removed	0.8247	$y = (5.289 \times 10^{101}) e^{(-5.6556 \times 10^{-3})x}$
MP7	none	0.8417	$y = (1.0771 \times 10^{126}) e^{(-7.0013 \times 10^{-3})x}$
MP8	none	0.8347	$y = (4.9177 \times 10^{123}) e^{(-6.8728 \times 10^{-3})x}$

Table 2.3.1. Models developed to predict the radiation dose at MP1-MP8 beyond October 2012

 $y = \text{dose rate } (\mu \text{Sv/h}), x = \text{time (in days since } 00/01/1900)$

3 Site Characterisation

3.1 Radiation Mapping

Since the approximate location of both the Stations and Monitoring Posts at Fukushima are documented by TEPCO, the radiation levels could be mapped. A grid was overlaid with the station location map to determine x-y coordinates for the measurement locations. The origin was set at the epicentre of the disaster, where the crippled reactors leaking nuclear fuel are located, and the position of the stations calculated relative to Unit 1 (see Figure 3.1.1). With coordinates set for the measurement locations on the x-y plane, the radiation level at those locations can be represented in the z direction. This allows radiation across the site to be depicted in 3D surface plot and easily visualised. It is apparent from Figure 3.1.2 that initially (01/05/2011) radiation levels descended fairly steadily from the centre of the site outwards. However, in the 5 years since the accident, clean-up and decay has most significantly reduced radiation in the intermediate zone. Levels at the periphery are still noticeable but

low, and doses are dangerously high near the reactors. To further quantify radiation levels and understand the effect and need for soil removal, the depth distribution of radiation in soil must be considered.



Figure 3.1.1. Mapping coordinates showing Monitoring Posts (MP1-MP8) and Stations (S1-S7) with the origin centred at the reactor units and epicentre of the accident.



Figure 3.1.2. Radiation dose as a function of location soon after the Fukushima accident (*top*) and current levels (*bottom*).

3.2 Soil and Radiation Distribution

Since this study is concerned with soil removal, determining the depth of removal is critical to further calculations. Significant research has been focussed on determining the distribution of radionuclides through soil depths, in particular the distribution of caesium isotopes. Kato et. al. (2012) observed 86% of the total deposited radiocaesium in the upper 2.0cm of soil which reflects other models which also suggest an exponential distribution favouring the near-surface region (Matsuda et. al. 2015). As surface deposited isotopes, both from the initial explosion and atmospheric radiation, are the greatest sources of radiation in soil this is a fairly natural distribution, especially considering natural diffusion through the ground (Koarashi et. al. 2012). Some efforts have been made to further quantify this diffusion through caesium tracking but results suggest effects are negligible when ground works such as ploughing are not active at the site (Koarashi et. al. 2012, Kato et. al. 2012).

The high percentage of radiocaesium in the near surface region means top-soil removal is a viable method of reducing radiation hazards. It is generally agreed that based on caesium distributions, a 5cm top-soil removal is both a practical and effective initial removal depth. This investigation is interested in the removal of all radionuclides and thus assumes that other radioactive isotopes also conform with the well-established caesium distribution and

will be eradicated in the same top-soil removal. This is a reasonable assumption, particularly considering the abundance of caesium at Fukushima and its dominant contribution to radiation doses. In order to demonstrate this, the radiation doses at Fukushima Daiichi were combined with weighting factors established by the International Commission on Radiological Protection (ICRP) for different types of particle decay. The resulting breakdown is shown in Figure 3.2.1, illustrating that heavy ions and fission fragments are most prevalent in Fukushima dose rates and most harmful to human health, carrying a weighting factor of 20. Caesium-134 and caesium-137, are the primary caesium isotopes expected in fission and decay products, and their relative abundance is a direct reflection of half-lives of 2.06 years and 30 years respectively (Brookins 1984). Their dominance of radiation dose (>86%) means the exponential depth distribution of caesium is a reasonable model for combined radiation at Fukushima.



Figure 3.2.1. Percentage contributions of radioactive decaying particles. Absorption doses developed from the International Commission on Radiological Protection's (ICRP) revised weighting factors (2007). Elemental contributions calculated as ratios from Fukushima Daiichi readings (September 2016)

R3 The exponential distribution of radionuclides favouring the near-surface region means top-soil removal is the most viable and efficient elimination remediation method at Fukushima Daiichi.

3.3 Calculating Soil Removal

Based on the findings of Kato et.al (2012) and Yasutaka et.al. (2016), a relationship between the effectiveness of top-soil removal and the starting radiation dose for that area can be formulated (see Equation 3.3.1). Defined in a piecewise manner, the soil removal required to restore each zone of the Fukushima site must be evaluated in discrete steps. Variability also exists in the desired restoration level, which remains debated (Omoto 2013). Here the case for complete restoration and remediation is considered; that is reducing reducing each zone back to near background radiation or minimum detection levels ($\approx 0.037\mu$ Sv/h) (Harada et. al. 2013).

$$f(y) = \begin{cases} 0.66y & \text{for } y \le 1 \ \mu \text{Svh}^{-1} \\ 0.51y & \text{for } 1 < y \le 3 \ \mu \text{Svh}^{-1} \\ 0.53y & \text{for } 3 < y \le 10 \ \mu \text{Svh}^{-1} \\ 0.20y & \text{for } y > 10 \ \mu \text{Svh}^{-1} \end{cases}$$
(3.3.1)

Given that Section 2 established radioactive decay at the Fukushima site conforms with an exponential relationship, soil removal equations can be developed for a general exponential expression:

Equation for Station i radiation level (y) in μ Sv at time x (with model parameters A_i and p_i):

- without top-soil removal, $y_i(x) = A_i e^{-p_i x}$
- with 5cm top-soil removal on date x_1 , $y_i(x) = A_i e^{-p_i x} - K_1 A_i e^{-p_i x_1}$ where $K_n = \begin{cases} 0.34 & \text{for } A_i e^{-p_i x_n} \leq 1 \ \mu \text{Svh}^{-1} \\ 0.49 & \text{for } 1 < A_i e^{-p_i x_n} \leq 3 \ \mu \text{Svh}^{-1} \\ 0.47 & \text{for } 3 < A_i e^{-p_i x_n} \leq 10 \ \mu \text{Svh}^{-1} \\ 0.80 & \text{for } A_i e^{-p_i x_n} > 10 \ \mu \text{Svh}^{-1} \end{cases}$ • with 5cm top-soil removals on dates x_1 and x_2 , (3.3.2)
- with 5cm top-soil removals on dates x_1 and x_2 , $y_i(x) = A_i e^{-p_i x} - K_1 A_i e^{-p_i x_1} - K_2 A_i e^{-p_i x_2}$
- with 5cm top-soil removals on dates x_1, x_2, \dots, x_n $y_i(x) = A_i e^{-p_i x} - K_1 A_i e^{-p_i x_1} - K_2 A_i e^{-p_i x_2} - \dots - K_n A_i e^{-p_i x_n}$

Substituting A_i and p_i with the relevant model parameters for each station means the radioactive decay at each station can be plotted for no clean-up actions, as well as 5cm, 10cm, etc. soil removal. This generates pay-back style charts, whereby cost is substituted with the more immediately relevant radiation level. An example of one such payback chart is shown for Station 3 in Figure 3.3.1, demonstrating the clear benefit of a 5cm removal attaining the chosen base level of 0.037μ Sv/h quicker. Similar results for the remaining stations are summarised in Table 3.3.1. Whilst these charts are visual tools and powerful for qualitative statements, in order to conduct a quantitative analysis the earlier set of expressions must be reconsidered. Rearranging the general exponential equation means the date forms the subject of the expression, and can be determined as a function of the desired dose level.



Figure 3.3.1. Modified pay-back period charts balancing radiation levels with time to decay with and without removal

Equation to determine date (x) when the radiation level drops to y (μ Sv) at Staiton i (with model parameters A_i and p_i):

- without top-soil removal, $x_{i}(x) = \frac{1}{-p_{i}}[ln(y) - ln(A_{i})]$ • with 5cm top-soil removal on date x_{1} , $x_{i}(x) = \frac{1}{-p_{i}}[ln(y - K_{1}A_{i}e^{-p_{i}x_{1}}) - ln(A_{i})]$ where $K_{n} = \begin{cases} 0.34 & \text{for } A_{i}e^{-p_{i}x_{n}} \leq 1 \ \mu\text{Svh}^{-1} \\ 0.49 & \text{for } 1 < A_{i}e^{-p_{i}x_{n}} \leq 3 \ \mu\text{Svh}^{-1} \\ 0.47 & \text{for } 3 < A_{i}e^{-p_{i}x_{n}} \leq 10 \ \mu\text{Svh}^{-1} \\ 0.80 & \text{for } A_{i}e^{-p_{i}x_{n}} > 10 \ \mu\text{Svh}^{-1} \end{cases}$ (3.3.3)
- with 5cm top-soil removals on dates x_1 and x_2 , $x_i(x) = \frac{1}{-p_i} [ln(y - K_1 A_i e^{-p_i x_1} - K_2 A_i e^{-p_i x_2}) - ln(A_i)]$
- with 5cm top-soil removals on dates x_1, x_2, \dots, x_n $x_i(x) = \frac{1}{-p_i} [ln(y - K_1 A_i e^{-p_i x_1} - K_2 A_i e^{-p_i x_2} - \dots - K_n A_i e^{-p_i x_n}) - ln(A_i)]$

Based on these results, the earliest date radiation levels will return to base level, given a certain set of conditions, can be determined. Here, the 1st day of each year has been arbitrarily chosen as the date for soil removal. This however can be adjusted and the modelled redeveloped for other scenarios. Importantly, this allows the viability and efficiency of soil removal to be evaluated.

	Earliest	t Expected Date t	o Return to Back	ground Radiati	on Levels
	None	5cm removal (01/01/17) [Y/S]*	2nd 5cm rem	oval (01/01/18) [Y/S]*
Station 3	21/09/2030	12/04/2019	[2.2]	04/10/18	[0.2]
Station 3 (port)	09/12/2025	20/08/18	[1.4]	11/05/2018	[0.1]
Station 6	21/12/2039	13/09/2017	[4.4]	-	
Station 7	24/05/2033	30/05/2019	[2.8]	-	
MP1	24/11/2025	10/10/2019	[1.2]	18/12/18	[0.2]
MP2	16/08/2016	-		-	
MP3	19/06/2021	30/06/2018	[0.6]	27/03/18	[0.1]
MP4	27/06/2021	04/07/2018	[0.6]	-	
MP5	05/07/2017	-		-	
MP6	24/12/2014	-		-	
MP7	08/10/2014	-		-	
MP8	07/10/2014	-		-	

Table 3.3.1. Effect of removal compared to natural decay accompanied by payback period value known as impact factor

* [Y/S] = Reduction in years / depth of removal = Soil removal impact factor

3.3.1 Impact Factor

Table 3.3.1 includes an impact factor (Y/S) aimed at quantifying the efficiency of a top soil removal under given conditions, much like a pay-back period does when considering business ventures. It is immediately apparent that the initial 5cm removal has a more drastic effect across all non-zero stations than the secondary removal. This reflects the research showing radiation is exponentially distributed and further validates top-soil removal as an effective remediation strategy. Here any top-soil removal which generates an impact factor greater than 0.46 (i.e. a removal which reduces years by at least half the depth removed in cm) is considered a viable operation. This is based on taking 10% of the longest time to radioactive decay (2039 - 2016 = 23 years $\rightarrow 2.3$ years) as the minimum reduction time of interest to TEPCO. 10% was chosen arbitrarily, but optimisation studies show anything less than a 10% time improvement is typically not considered worth the cost of improvement (Boyles et. al. 2012). Then since 5cm has been established as the removal depth, the minimum viable Y/S = 2.3/5 = 0.46.

3.3.2 Limitations at High Doses

Whilst the majority of measuring stations across the Fukushima precinct exhibited exponential decay as was modelled in Section 2, those stations closest to the damaged reactors maintained dangerously high doses throughout the 5-year period considered. These near constant dosages therefore require slightly different mitigation. For the scope of this report, top-soil removal is considered an applicable process even at greater depths. Iteratively applying the removal equation (see piecewise definition in Equation 3.3.1) means the depth of removal can also be determined in these epicentre areas. However, it is important to note that in these immediate regions, the leaking nuclear fuel will need to be removed to prevent further soil contamination and the need for further removal. The results at the remaining stations are summarised in Table 3.3.2 along with the required soil removal from the initial stations, as determined by the impact ratio (Y/S).

Table 3.3.2. Model predictions of required soil removal depth (cm) at Fukushima measuring stations

S(2)	S((3)	S(7)	S(4)	S(6)	S(1)	S(5)	MP1	MP2	MP3	MP4	MP5	MP6	MP7	MP8
65	5	5	5	5	60	5	70	65	5	0	5	5	0	0	0	0

3.3.3 Removal Volume

In order to quantify the magnitude of remediation required at Fukushima, the results of the radioactive decay modelling, site mapping and depth distribution were combined. Figure 3.3.2 illustrates the depth of soil removal required across the site as a 3D surface. The volume of this removal was estimated using the simple volume of a pyramid, and confirmed using a more accurate numerical double integration. Based on past radiation data, the current volume of soil removal required for the immediate area surrounding the Fukushima Daiichi plant totals 778 710 m³. Given that the longest natural decay of the soil removed is 2039 (from Table 3.3.1), storage sites must be sufficient to house this waste at least until 2039. Having established removal volume through a detailed analysis the material impact and cost of such an operation can be determined.



Volume Estimation (Pyramid) $V = \frac{1}{3} lwh = \frac{1}{3} (1)(3)(7 \times 10^{-4})$ $V = 7 \times 10^{-4} \text{km}^3 = 700\ 000\ \text{m}^3$

Volume Approximation (Numerical Integration) $V = \iint R(x,y) \, dx \, dy \approx (\sum \text{Removal depths}) \, dx \, dy$ $V = 7.7871 \times 10^{-4} \text{km}^3 = 778 \, 710 \, \text{m}^3$

Figure 3.3.2. 3D volume demonstrating soil to be removed

R4 The decommission operation must include the preparation of storage sites within a 15km radius, capable of storing 778710m³ of waste, at minimum until 2039.

4

Material and Energy Impact

4.1 Removal Process

Based on the radiation data recorded by TEPCO and the modelling developed here, the effectiveness of top-soil removal has been evaluated. This does not however account for the energy, raw material and human resources the clean-up operation will consume. These factors are most important in achieving clean up goals and restoring the Fukushima region as they dictate both the time frame and cost of the project. Researchers generally agree that prior to top-soil removal a wetting agent is critical to preventing further spread of radioactivity (Yasutaka & Naito 2016) and that plastic is sufficiently gas tight and affordable for isolation (Nakano & Yong 2013). Thus soil handling can be broken into a process of (1) preparing the area by wetting down, (2) excavating soil, (3) encapsulating soil in plastic bags, (4) transporting bags to a storage site, (5) ongoing storage and maintenance and (6) decommissioning once radiation has dropped below safe levels. In order to evaluate the raw materials required to both remove soil and also store it over the decay phase in these storage facilities, a detailed material audit was completed.

4.1.1 Raw Material Consumption

Plastic bag:Modelled as a $1m^3$ capacity cube \rightarrow SA = $6 \times 1m^2$ (calculations all per m^3 soil removed)
At approximately 2mm thick, as suggested in Yasutaka & Naito (2016), volume of material is:
 $V = Ah = 6(0.002) = 0.012m^3$
 $m = V\rho = 0.012(917.5) = 11.01kg$ ($\rho = 0.9175g/cm^3$ from Beswick & Dunn (2002))

Wetting agent:	Estimated as $3L/m^2$ of exposed soil (similar to most pesticides): Assuming a concentrated chemical: $\frac{1}{6}$ agent, $\frac{5}{6}$ water $\rightarrow 0.5L/m^2$ of wetting agent, $2.5L/m^2$ of water To convert from surface area to a per volume basis, consider a 5cm top-soil removal: $\frac{V}{h} = A$ $\frac{1}{0.05} = 20m^2$ $20m^2$ land area $\rightarrow 1m^3$ soil removed $\rightarrow 10L$ of wetting chemical per m ³ of soil removed, 50L of water per m ³ of soil removed							
Plastic top sheet:	Cover sheet to further isolate contaminated soil from the atmosphere and environment Assuming bags are stacked 5 high (a common level in many schematics), $\rightarrow 1\text{m}^2$ plastic sheeting per 5m ³ soil stored, or 0.2m^2 plastic sheeting per 1m ³ soil stored Regulations stipulate 2 × 1.5mm thick sheets of high density polyethylene (HDPE): $\rightarrow V = 0.2(2 \times 0.0015) = 0.0006\text{m}^3$ (per m ³ of soil)							
Plastic bottom sheet:	As above, 0.2m^2 plastic sheeting per 1m ³ soil stored, with 3 × 2mm sheets: $V = 0.2(3 \times 0.002) = 0.0012\text{m}^3$ m = 0.0012(964) = 1.1568kg							
Rough sand:	Based on schematics, approximately 1m^3 of sand required per 10m^3 of soil removed to create banked, earthquake proof structure: $m = 1(1281) = 1281\text{kg for } 10\text{m}^3$ ($\rho = 1.281\text{g/cm}^3$ from Callister et. al. (2000)) $m = 128\text{kg per } 1\text{m}^3$ of soil removed							
Diesel fuel:	Typical haulage trucks have a 4 tonne capacity (Yasutaka & Naito 2016), $\rightarrow 0.25$ trucks per 1m ³ $0.25 \times \text{fuel consumption} = 0.25 \times 3.25 = 0.8125 \text{L/km}$ (3.25L/km from Komatsu (2010)) Given that most storage sites are approximately a 30km return trip (Yasutaka & Naito 2016): V = 0.8125(30) = 24.4 L							

4.1.2 Energy and Mass Flow of Process

The results of the material consumption calculations in conjunction with additional energy inputs are presented in the form of an energy mass balance (Figure 4.1.1) which describes the soil removal process required for radiation clean-up. Soil removal, like most clean-up processes, is a resource intensive task and in the context of a system provides very limited useful outputs. This is a well established notion in oil spill clean-ups (Etkin 2000) and the consumption of materials and energy are considered necessary simply to prevent further environment harm or health risks. Figure 4.1.1 does however highlight several areas for waste reduction. The radioactive decay of elements does result in the emission of energy (typically heat) which albeit low, could be harvested to at least power monitoring instruments at the storage site. The viability of such an operation is unproven and the simplicity of storage sites as isolated decay sites is considered preferable by some communities (Nakano & Yong 2013).

A more promising avenue for material and energy reduction is the reprocessing of nuclear material. Research into extracting radionuclides from soil is ongoing and has the potential to radically reduce the volume of soil removed (Satou et. al. 2016). Even if radioactivity can only be concentrated, any reduction in the volume of soil to be stored will significantly reduce material use. Only preparation and excavation are required for reprocessing, reducing consumption to a small mass of diesel, water and a wetting chemical. In contrast, storage consumes in excess of 12kg of plastic per cubic metre of soil. Furthermore, plastic is the greatest contributor to the overall embodied energy of the system, consuming the most energy in production and transport to Fukushima. Concentrating radioactive waste would greatly reduce both material and embodied energy consumption as well as the space required for natural decay over approximately 2 decades.



Figure 4.1.1. Energy-mass flow depicting the soil removal and storage process encompassing the entire life-cycle flows into and out of the system, including raw material and energy quantities.

Due to the radioactive nature of Fukushima soil, the plastic used during encapsulation cannot be recycled by traditional methods or reused in other sectors (Yasutaka et. al. 2016). The cost of manufacture and transport is consumed by the clean-up operation, and any material remaining after decommissioning is designated for incineration, storage and is ultimately deposited to landfill. Thus, a focus on reducing plastic consumption, or substitution of several layers of low-density polyethylene with fewer, thin sheets of high-density polyethylene should become a priority of the Fukushima clean-up. The embodied energy of raw materials also closely correlates with cost and whilst this has become a forgotten element in the Fukushima clean-up, it is further evidence of the potential for a reduction in soil volume to impact material and energy consumption, as well as reducing economic ramifications.

Table 4.1.1. Material audit of soil removal process as described in Figure 4.1.1 (all measurements per m^3 of soil removed) and typical embodied energies, cost from Mo (2011), Gilbert (2009), Salih (2013) and Franklin (1991)

Descriptor	Material	Quantity	Embodied Energy	Raw Material Cost
Fuel	Automotive diesel	20.3kg	54.9MJ/kg $ ightarrow$ 1114MJ	121 JPY/kg ightarrow 2460 JPY
Wetting agent	Surfactant (wetting)	10.0kg	$\rm 88.5 MJ/kg \rightarrow 885 MJ$	$\rm 2620 JPY/kg \rightarrow 26200 JPY$
Water	Bore, river water	50.0kg	$0.01 \text{MJ/kg} \rightarrow \textbf{0.5MJ}$	on-site
Plastic bag	Low-density polyethylene (LDPE)	11.0kg	$103 \text{MJ/kg} \rightarrow 1133 \text{MJ}$	$727 \text{JPY}/\text{kg} \rightarrow \textbf{8000JPY}$
Retainer	Rough sand	130kg	$0.10 \text{MJ/kg} \rightarrow \textbf{13.0MJ}$	on-site
Plastic btm. sheet	High-density polyethylene (HDPE)	1.20kg	$103 \text{MJ/kg} \rightarrow \textbf{123.6MJ}$	$800 JPY/kg \rightarrow 960 JPY$
Plastic top sheet	High-density polyethylene (HDPE)	0.58kg	$103 \text{MJ/kg} \rightarrow \textbf{59.7MJ}$	$\rm 800 PY/kg \rightarrow 464 JPY$
	Materials used to remove and store $1m^3$ of soil		Total 2786MJ	38080JPY

Investigate reprocessing nuclear material to concentrate radioactivity in a smaller volume, thus reducing raw material consumption and embodied energy of the removal process.

R5

5

Outcomes, Cost and Alternatives

5.1 Life-Cycle Cost

Eliminating the health risks posed by radioactive material remains the priority of the Fukushima clean-up, however the cost of soil remediation is relevant in demonstrating the magnitude of the operation, and can be estimated based on the modelling and material impact factors developed previously. Summing the cost of raw materials for disposal and storage, estimating the human resources required to carry out the operation, and including the ongoing and decommissioning costs calculated by other researchers (Yasutaka & Naito 2016), indicates a total clean-up cost of 105 680 JPY/m³. Inflation factors haven't been included in this figure, primarily because the majority of these costs are incurred at start-up, and thus in AUD this figure equates to 1350AUD at current exchange figures (September 2016). Ultimately the cost of this operation is incurred by the Japanese government and is absorbed by the profits of the entire nuclear industry. This study has established that in the immediate areas surrounding the Fukushima Daiichi Nuclear Power Plant alone, soil remediation will cost in excess of 82 billion JPY (≈ 1.0 billion AUD).

Table 5.1.1. Life-cycle cost

Cost Type	JPY/m ³
Raw materials	38080
Labour*	1600
Ongoing	
Temporary site	20000
Interim site	16000
Decommissioning of site	30000
TOTAL	105680

*Labour estimated from a quote received utilising a truck driver, excavator operator, ground clearer and supervisor

R6 Funds should be prepared for an 82 billion JPY operation to remove soil in the immediate Fukushima precinct, remediate environmental damage, reduce health risks, and restore land for farming purposes.

5.2 Alternative Actions

The reprocessing of nuclear material has already been discussed but significant progression of technology would be required for this to handle radioactive soil exclusively (Satou et. al. 2016). A more common suggestion which has been employed at the highest activity areas of Fukushima is concrete pouring over contaminated soil (Yasutaka & Naito 2016). This employs the weaker risk control measure of isolation (compared to removal enacting elimination) and concedes that these areas will never become inhabitable. However, considering the combined economic, energy and material cost of removal, this may ultimately prove a viable option. The consumption of this process itself is beyond the scope of this investigation but is certainly a direction for future research and important in developing a comparison. The results of this analysis do however demonstrate that despite the natural decay of radioisotopes, the exponential rate means the last phase of decay is exceptionally drawn out, and any system is hindered by the immense storage time required and large volume of weak but nonetheless radioactive soil.

5.3 Conclusion

Based on historic radiation dose levels throughout the Fukushima Daiichi plant, natural radioactive decay was determined throughout the immediate precinct. These predictions, in conjunction with published top-soil removal data allowed a model for removal to be developed and the total volume of soil destined for removal to be estimated. Considering the raw material and energy consumption per unit volume of soil removed, indicated key areas for improvement, and highlighted the magnitude of soil mitigation required in the most immediate areas of Fukushima alone. The investigation confirmed that (R1) radioactive soil is a potent risk in need of remediation through careful engineering design, and (R2) TEPCO data serves as reliable source for modelling purposes. (R3) Top-soil removal was established as the most effective elimination method but will require (R4) storage sites capable of storing 778 710m³ of soil beyond 2039, (R5) necessitating improved processing methods. (R6) The estimated 82 billion JPY cost of soil removal alone demonstrates the resource intensive nature of remediation and demands the investigation of other methods. Ultimately a combined system of removal, reprocessing and storage is likely critical to efficiency in the Fukushima clean-up operation and restoring the precinct to pre-accident conditions.

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