

Response to Editor Comments (indented paragraphs in bold)

Comments to the Author: esurf-2020-72, The effects of topography and soil properties on radiocesium concentrations in forest soils in Fukushima, Japan

General response from the authors

We greatly appreciate the detailed review and comments provided and the patience exhibited by the editorial team. With these detailed comments and suggestions, we undertook a major rewrite of the paper.

Major changes include the following:

1. The title of the paper has been revised to more accurately represent the material presented. It is now entitled “Assessing the Effect of Topography on Cs-137 Concentrations within Forested Soils due to the Fukushima Daiichi Nuclear Power Plant Accident, Japan.”
2. The Introduction now includes information about the nuclear accident, a stronger justification for the current study, and clearly defined objectives for the current paper that are strongly aligned with the results presented.
3. The Methods section has been rewritten. All methodological information on sampling, soil and radiation analyses, DEMs, and statistical analyses have been consolidated into a single section and presented in a logical order.
4. All primary data collected within this study have been included in an Appendix.
5. Several plots and figures were removed, revised, and consolidated, and unnecessary analyses and discussions were deleted. The total number of figures were reduced from 26 to 13.

In the sections below, the comments from the referee and editor are shown in italics, and the authors' responses are shown in normal text and indented.

This manuscript received three reviews and I have read it in detail myself. Two reviewers were broadly supportive, but reviewer two was sceptical, stating that the motivation of the study was poor. Reviewer one, was positive, though makes a number of important points, and doesn't comment explicitly on the statistical methods. Reviewer 3 comments critically about the vertical resolution in relation to the samples collected.

Whilst this is undoubtedly an interesting problem and data set I have to agree with reviewer two that the motivation for the study would need to be much more clearly conveyed if this is to be published. An interesting data set and statistical analysis is not sufficient for publication. The bar is high in terms of how this needs to be presented if this is to be understood by the broad audience of ESURFD. In the authors response, I don't think they addressed sufficiently the comments of R2.

We now have rewritten the Introduction to include more background information on nuclear accidents, a stronger justification for the overall goal of the research that focuses on the effects of topography on Cs-137 concentrations, and clearly defined objectives of the current paper that are in complete alignment with the results presented. This scientific motivation and research goals also have been included in the Abstract and Conclusions, where appropriate.

It is not reasonable to assume that readers will be familiar with all the statistical methods as well as the radiocesium literature.

We have provided a more complete description of the statistical models employed, and we have expanded the presentation and discussion of model results and their application herein. We then connected the model results to the overall goal of the paper, which is the effect of topography on Cs-137 concentrations.

In summary, this manuscript takes a statistical approach to determine whether geomorphological parameters influence the accumulation of radiocesium following a major nuclear disaster. A number of samples were collected over a 3 year period following the disaster from a relatively restricted area, over a relatively narrow range in altitude. The radiocesium was determined and a number of statistical models are used to test whether there are links with key topographic features such as elevation, slope, curvature and vegetation type. It is never really laid out to the reader why ¹³⁷Cs might correlate with such topographic parameters (the list on pp17 is the best but it should have come much sooner). What are the underlying physical/chemical/biological mechanisms (they get lost in the text and should be stated right at the beginning). On the one hand altitude might relate to the original deposition of ¹³⁷Cs (orographic effect), and vegetation type (and elevation) might relate to physical/biological/chemical processing of ¹³⁷Cs post deposition. Why should slope and curvature matter (this is briefly stated but not elaborated)? If these are the key parameters that control the accumulation (retention) of ¹³⁷Cs in the environment then some clear figures with the data are required to show this both in an individual and multivariate manner. The current figures do not do a sufficient job of conveying a clear message from the data and are set out in a disorganised manner. There are too many figures for the message that is being conveyed. There would be clearer, simpler more concise and convincing methods to elucidate a complex yet fascinating dataset.

We now provide a literature review and a theoretical framework for the topographic indices examined here and their potential impact on Cs-137 concentrations in soils at and just below ground surface. We rewrote the Introduction and Methods sections to provide this important context. In addition, the number of figures has been reduced significantly, and the primary data collected have been included as an Appendix in an effort to present a clearer and more concise reporting of the research.

There are a series of major issues that would need to be addressed if this were to be considered further for publication. Based on the author response to the initial reviews I'm not convinced that such a whole-scale rewrite is possible. There are organisational, methodological and grammatical issues which must all be addressed. As is, this does not read like a polished paper, but more like a detailed thesis or specialist report. Unfortunately, in the version I read, the figure numbering appears to have gone wrong which doesn't help. It may be, that it would be more appropriate in a specialised journal.

The paper has undergone a complete rewrite. While the results remain the same, we now provide much greater justification for the work, clearer separation of methods and results, expanded discussion and context for the methods employed, and a reduced number of figures to better focus the presentation and interpretation of the results.

Specific comments (not exhaustive)

1. Generalized additive models (GAM)

The statistical models (for example the Generalized additive models (GAM)) seem complex for what the data shows. The GAM method is not well explained. What exactly is being minimised to generate the residuals? I'm not sure the text and figures do justice to the data? A large number of the figures (and there are too many) show deviance or residuals. It is difficult to ascertain from these figures the validity

of the proposed covariation or not. I don't think it is sufficient to say that the "gam.check ()" function was used. If readers aren't familiar with R or this particular function in R, this will convey very little.

We now include a greater explanation of GAMs, their mathematical basis, the approach used to create the models, and a description of the indices used to assess model performance. The phrase "deviance explained percentages" is used, which is the generalization of R^2 in the GAM algorithm. We also explained the gam.check() function employed in the R-package.

There is no obvious or particularly intuitively relationships between 137-Cs and topographic parameters. For example, in section 6.2 accumulation patterns on a simple representative hillslope, 3 figures are shown. The first appears to show the relationship between elevation and 137-Cs. It is clear that there is no simple relationship. Perhaps this is why a complex GAM is required. What is the trend line on this diagram? What is the simple representative hill slope? The second figure appears to show slope vs elevation, but how does this relate to 137Cs?

It is precisely because there is no obvious relationship between Cs-137 concentration and topographic indices that we explored statistical models. GAM is a statistical detection tool to assess quantitatively relationships or trends amongst parameters that other regression methods fail to address. The graphs noted by the referee have been updated and simplified to improve the communication of the results and their interpretation.

Perhaps clearly subheadings should guide the reader through the data analysis in a more systematic way such as:

- I) Elevation*
- II) slope*
- III) soil water content*
- IIII) density*

In our rewrite, we have revised all headings and reorganized all results and discussions to greatly improve the logical flow of the paper. In the current version, we now discuss the influence of each topographic and soil property parameter on Cs-137 concentrations. This section is intended to provide a clearer and stronger interpretation of the GAM results and the impact of topography on Cs-137 concentrations.

At the moment, the data description and discussion appear to be driven by the statistics (four tests), rather than clearly testing a hypothesis with a clear physical or geomorphological basis. If the 4 tests were retained, they should be laid out much more clearly so the reader can follow. An example of how I was unable to follow even the Single parameter GAM results is given below:

We have eliminated the Tukey test (which tested the significance of vegetation cover types and slope aspects) and the spatial prediction aspects of the paper, so that our current effort remains entirely focused on topography. While we recognize that vegetation may play an important role in modulating Cs-137 concentrations and that spatial predictive analytics could be of interest, these previous discussions did not bring added value to the paper. In addition, we now provide all primary data collected in this study in an Appendix, to further inform the reader.

Page 28:

As I understand section 6.2, it is examining individual parameters as a controlling factor on 137-Cs. It is a

pity that it is not possible to indicate the lack of correlation (as I understand it) with a figure. However, what I really don't understand is the following sentence: "None of the single parameters in either table returned deviance-explained percentages above 36 % (which was found via a model including water content)."

I don't understand how the water content is being modelled at this point. How the single parameter GAM works has not been well enough explained. Being single parameter, I had clearly wrongly assumed that it meant it was dealing with one parameter at a time.

For simplicity and clarity, we presented the GAM results in a table rather than graphically, and we also sought to improve the structure and context of the supporting discussion. The sentence in question has been revised accordingly.

2. Section 4.1 and 4.2: DEM work

This section seems to deal with extracting key parameters from the different resolution DEMs. However, it also seems to mix in 137Cs measurements (line 281). This doesn't seem like the right place to know the units of 137Cs. The work on the DEM resolutions is clearly important, but equally it is a distraction from the main message of the manuscript. Can section 6.3.1 move to an appendix? Equally, the organisation seems to be confused. Later on in the manuscript, Fig 15 (Slope degrees of sampling points along elevation (m)) seems to be entirely related to the DEM work and have nothing to do with radiocesium. There are clearly major organisational problems.

We restructured the article in the following ways. All methodological information regarding the DEMs and the topographic indices are now presented in the Methods section, which also includes additional information for the selection of these indices in the context of landscape processes. We include all primary data collected in this study, including the topographic indices extracted from the DEMs, now compiled as a table in an Appendix. Lastly, we have reorganized the presentation of the results to strengthen the narrative and provide a logical sequence for the topics introduced.

3. Figures

There seems to be a major problem with the numbering of the figures.

Figures should be understandable just by looking at the figure and the caption. For example, Fig 12 (pp36). What are the solid red lines. In the caption, it refers to top row graphics and bottom row graphics. What are these?

First, we reduced by one-half the total number of figures in the paper. Second, we have revised many of the remaining figures and their captions to improve their graphical presentation and ease in interpretation. Figure 12 noted here has been deleted.

Fig 4: It is a shame that there is nothing higher resolution available than this. As is, it is not very satisfactory. Could this not be incorporated into fig 5, plotting the sample locations in 3 different colours (if I have understood the figures)?

We eliminated several figures, and now present a single image (Fig. 3) to demarcate the sampling locations clearly and efficiently.

On Fig. 15, what are the red dots. It is not clear from the Fig or the caption.

The red dots have been changed to numbers, which refers to the four samples with the highest Cs-137 concentrations.

Line 432: "In Fig. 13, both trend lines of Cs-137 in Bq m-2 values show upward trend along elevation." Fig 13 appears to show depth vs 137Cs and COV.

The revised figures (Figs. 7 and 8) now show Cs-137 distributions plotted against elevation.

Figure 3. An aerial views of the study site, facing northwest. This would be fine in a Phd thesis, but is not appropriate in a paper. In addition, it does not appear to be an aerial view as is stated.

This figure has been removed.

Figure 6: The low water content appears to contradict the 100% statement (referred to below). There appear to be a large number of outliers on the high side. Would this be best shown as a histogram? It is hard to visualise how much data is in the box.

We converted the tables to boxplots, now shown as Fig. 6.

Figure 8: Nice to see a photo, but is it needed? What does the photo convey scientifically? This would be fine in a thesis, but not a paper.

This figure has been removed.

Can figures 9 and 10 be combined with fig 8 that comes after 9 and 10? Or combine 8, 12 and 13, the interesting depth figures.

To improve paper clarity, we combined several plots into single figures. Figs. 8 and 9 are now presented as Fig. 4, Figs. 10, 11, and 12 are now presented as Fig. 5, and Figs. 13 and 14 are now presented as Fig. 7. We also modified these graphs to improve their interpretation by the reader.

Fig.17: Hard to know what it means.

This figure has been removed.

Can Fig. 9, 10 and 20 be combined? (Pp30 and 31)

Where possible, we have combined many plots into single figures, which was done to improve the clarity and presentation of the results.

The Fig on pp35 appears to be very useful, comparing model predicted 137Cs with data. A pity there is no fig number or caption.

As it happens, this figure has been removed.

4. Writing style and conciseness:

There are numerous examples where the writing style does not have the conciseness required for a scientific publication. Avoid phrases like "This section displays" and make a point directly.

Other examples (non-exhaustive): "Literature review" Seems like an odd heading for a scientific paper. Sounds more like a thesis. "When driving into the study site region, visitors see hills and mountains covered with forests. Winding, narrow roads connect". Not appropriate for a concise scientific paper. "natural" or "native". Choose one option

We carefully edited the paper to address any lapses in language and word usage. All changes were applied as suggested.

Line 140: Land use comments: This is not the right place for this comment. This section is about the study site. If and use matters, then include it. If not, then say it doesn't

Amended as suggested

Line 157: perhaps change to "to confirm anomalies observed in previous years..."

Amended as suggested

Line 159: change to "but mostly on the south-west side due to accessibility"

Amended as suggested

Line 190: Particles larger than 2mm were removed with a sieve

Amended as suggested

Line 198: The unit of measurement is the kiloelectron volt (KeV), equivalent to the kinetic energy gained by an electron falling through a 1 volt potential.

Amended as suggested

Line 215: "Approximately less than". Just less than

Amended as suggested

Line 217. Missing full stop and space.

Amended as suggested

Line 226: "Only some of the samples were tested for texture because of time and human resource constraints during the limited lengths of the first author's stays in Japan." Remove. Informal detail not needed.

Amended as suggested

Other

Line 433: Camelback. This is not scientific terminology. The manuscript goes to a great effort to produce a set of descriptive statistics to define relationships between parameters. This shouldn't be undermined by colloquial vocabulary.

Amended as suggested

Center of gravity (COG) depth (cm): What is this?

This term has been deleted.

"Migration head depths" are not defined. This appears to be first mentioned on line 413.

We now clearly define this parameter, shown graphically in Fig. 7.

Soil properties are discussed, but not in great detail. What about the cation exchange capacity of the soils for example that Cs has a very high affinity for. Presumably the cation exchange capacity is thought to be constant in these soils. Is this also true with depth. It seems to me that one major variable that has been overlooked in this analysis is the chemistry. How much of the Cs is partitioned between aqueous Cs and the soil?

McHenry and Ritchie (1977) stated that CEC was an important factor to consider for Cs-137 modeling. There are reasons why the soil chemistry topic was not incorporated into this study.

1. We did not have publicly available soil chemistry data for the study site soils.
2. We did conduct pH, CEC, and C/N ratio tests on a very limited number of samples selected for texture analysis and other research purposes (not included in this article). But the total number of samples tested was relatively low due to a lack of time and resources. We do believe the data presented herein was sufficient for the purpose of the objectives of the current paper.
3. It should be noted that soil chemistry data might not explain the Cs-137 concentration patterns directly. For example, CEC becomes affected by the percentages of organic matter in the soils and CEC itself might not linearly explain Cs adsorption rate (Mensah, et al., 2020).
4. There is ample literature that has incorporated chemical properties with Cs-137 analysis (Matsunaga, et al., 2013, Mori, et al., 2015, Parajuli, et al., 2013, Takahashi, et al., 2015).

How can the average water content > 100%. This was also asked by reviewer 1. I don't understand the author response. How can the moisture be in excess of 100%. I must be mis-understanding the definition.

It is now made clear that we report 'mass water content,' which can achieve values higher than 100%.

References for this note:

Matsunaga, T., Koarashi, J., Atarashi-Andoh, M., Nagao, S., Sato, T. and Nagai, H.: Comparison of the vertical distributions of Fukushima nuclear accident radiocesium in soil before and after the first rainy season, with physicochemical and mineralogical interpretations, Science of the total environment, 447, 301-314, doi:10.1016/j.scitotenv.2012.12.087, 2013.

McHenry, J. R. and Ritchie, J. C.: Physical and chemical parameters affecting transport of ^{137}Cs in arid watersheds, *Water Resources Research*, 13, 6, 923-927, 1977.

Mensah, A. D., Terasaki, A., Aung, H. P., Toda, H., Suzuki, S., Tanaka, H., Onwona-Agyeman, S., Omari, R. A. and Bellingrath-Kimura, S. D.: Influence of Soil Characteristics and Land Use Type on Existing Fractions of Radioactive ^{137}Cs in Fukushima Soils, *Environments*, 7, 2, 16, 2020.

Mori, K., Tada, K., Tawara, Y., Ohno, K., Asami, M., Kosaka, K. and Tosaka, H.: Integrated watershed modeling for simulation of spatiotemporal redistribution of post-fallout radionuclides: application in radiocesium fate and transport processes derived from the Fukushima accidents, *Environmental Modelling & Software*, 72, 126-146, 2015.

Parajuli, D., Tanaka, H., Hakuta, Y., Minami, K., Fukuda, S., Umeoka, K., Kamimura, R., Hayashi, Y., Ouchi, M. and Kawamoto, T.: Dealing with the aftermath of Fukushima Daiichi nuclear accident: decontamination of radioactive cesium enriched ash, *Environmental science & technology*, 47, 8, 3800-3806, 2013.

Takahashi, J., Tamura, K., Suda, T., Matsumura, R. and Onda, Y.: Vertical distribution and temporal changes of ^{137}Cs in soil profiles under various land uses after the Fukushima Dai-ichi Nuclear Power Plant accident, *Journal of environmental radioactivity*, 139, 351-361, doi:10.1016/j.jenvrad.2014.07.004 2015.