REVIEW OF FAIREWINDS STUDY

Introduction

A paper, entitled "Radioactive microparticles related to the Woolsey Fire in Simi Valley, CA", by M. Kaltofen, M Gundersen and A. Gundersen, was published in the Journal of Environmental Radioactivity on October 8, 2021.[1] The paper is provided as an appendix to this report and is reviewed below.

The key conclusions of the Fairewinds study, as stated by the authors, are,

- *"Data did not support a finding of widespread deposition of radioactive particles."* (Abstract, page 1).
- *"Given the low percentage of dust particles ultimately found to contain significant activity …"* (page 2)
- *"None of the radioactive microparticles detected in the study included any form (radioactive or stable) of cesium."* (page 5).
- *"The radioactive particles detected in this study are not definitively natural or industrial."* (page 7).
- *"A significant majority of samples (97% of 360 samples) collected in the study zone registered radioactivity that matched existing area background levels."* (page 7).
- *"The nuclides identified as the sources of excess radioactivity in impacted samples were predominantly isotopes of radium, uranium, and thorium."* (page 8).

[1] M Kaltofen et. al., "Radioactive microparticles related to the Woolsey Fire in Simi Valley, CA", https://pubmed.ncbi.nlm.nih.gov/34634531/ (Accessed 10/26/2021)

Thorium Monazite Microparticles

The authors introduce a new term, "microparticles", in place of "dust" or "particulate matter (PM)" implying that something new has been discovered.[2] The authors characterize these microparticles as *"containing sufficiently high concentrations of radioactive elements to exert an internal radiation dose if the microparticle is inhaled."* The implication is that these microparticles are a greater airborne radiological hazard than dust (containing naturally occurring or anthropogenic radionuclides). No data is provided to quantify the *"high concentrations*" within microparticles. Furthermore, since no airborne activity measurements were taken (with associated volume measurements) the study could not estimate potential airborne activity or associated internal dose from these microparticles.

The authors identify the naturally occurring mineral, monazite, as a common source of thorium (pages 2, 6, and 7). Indeed, discussion of off-site detections of thorium in microparticles always describe them as thorium monazite (Table 4).

- *"All thirteen radioactive microparticles contained percentage thorium concentrations as thorium monazite (Figures 4-6)"* (page 7).
- *"The radioactive microparticles detected in this study are not definitively natural or industrial. Notably, however, the most significant number of monazite particles were detected at the SSFL fence-line, evidencing that, whether industrial or natural, the Santa Susana Field Laboratory or its exposed bedrock is a potential source of the thorium monazite particles detected in this study"* (page 7).

Any potential thorium contamination originating from SSFL operations would have been either originally refined thorium-232 metal and/or various thorium isotopes generated as decay products of nuclear fuel made up of uranium-238, uranium-235 or thorium-232. If released to the environment these isotopes could potentially form oxides or salts, including nitrates,

[2] Microparticles are particles between 0.1 micron and 100 microns in size. This wide range therefore encompasses typical measurements of airborne particulate matter, PM 2.5, and PM 10. The unit, micron is the same as the unit, micrometer.

carbonates, sulphates, phosphates, etc., through common chemical reactions. However, thorium could not form complex minerals with cerium, lanthanum, phosphorus, neodymium, silicon, and calcium. Such a process requires a geological timeframe. Monazite is formed during the crystallization of igneous rocks and during the metamorphism of clastic sedimentary rocks.[3] Sedimentary rocks include sandstone which are common outcroppings in SSFL and neighboring Chatsworth and the Simi Hills. When these rocks weather, monazite is one of the more resistant minerals and becomes concentrated in the weathering debris. Contrary to the authors' uncertainty, thorium in monazite microparticles is naturally occurring, is a geologic age, and is not formed by SSFL nuclear operations or from potential environmental releases.

The authors are either unaware of this contradiction or intentionally ignore it. In a schizophrenic flip-flop, the paper switches back and forth between naturally occurring monazite and refined thorium used in some nuclear fuel at SSFL.

It appears that the authors, however unintentionally, have proven that the thorium in the thirteen monazite microparticles in seven samples (Table 4), and likely associated elevated alpha and beta measurements in a small fraction (3%) of locations is naturally occurring. The thorium in the monazite is unrefined and would have alpha and beta emitting progeny in secular equilibrium due to the geologic age of the monazite.

Sample 180 A collected at the maximum distance from SSFL (15 km) had the highest alpha count rate (Table 1) and the highest beta count rate (Table 2) of the entire dataset and was shown to have one of the highest monazites microparticle content (9.8%) (Table 4).

Alpha Count Rate

Of the 360 samples taken, 124 were bulk soil samples and 236 were "depositional" dust and ash samples taken using a 4-cm^2 adhesive lift-tape. These depositional samples are more commonly referred to as removable surface activity measurements or wipe tests. Additionally, similar lift

[3] "Monazite - Geological Occurrence of Monazite", Geology.com. https://geology.com/minerals/monazite.shtml (Accessed 10/27/2021)

tape samples were taken from the 124 homogenized bulk soil samples in the laboratory so that the total surface activity samples became 360. These were counted for removable alpha and beta surface activity. The study uses removable surface activity units of counts per hour per 4 cm^2 (CPHr per 4 cm^2). These units are converted in this review to the more conventional units of disintegrations per minute per 100 cm^2 (dpm per 100 cm^2).

counts per minute (cpm) per 100 cm^2 = counts per hour (CPHr) per 4 cm^2 * 25 / 60

4π alpha efficiency (cpm/dpm) for Ludlum 3030 = 35%[4]

dpm per 100 cm^2 = CPHr per 4 cm^2 * 25 / (60 * 0.35) = CPHr per 4 cm^2 * 1.19

Therefore, there is a close to 1:1 correlation between CPHr per 4 cm^2 and dpm per 100 cm^2.

The study reports the alpha count rate for the complete set of study samples to be 7.6 +/- 3.0 CPHr per 4 cm^2 (page 3 and Table 1). This is equivalent to a removable alpha surface activity of 9.0 +/- 3.6 dpm per 100 cm^2. Likewise, the upwind "background" samples had a removable alpha surface activity of 8.8 +/- 5.2 dpm per 100 cm^2. The authors correctly observe that the two means are not statistically different. What is more informative is comparing both the gross mean removable alpha surface activity of 9.0 dpm per 100 cm^2 and the net "background subtracted" removable surface activity of 9.0 - 8.8 = 0.2 dpm per 100 cm^2 to regulatory release limits. Both these metrics are well below the acceptable removable alpha surface contamination levels in USNRC Regulatory Guide 1.86[5] of 1,000 dpm per 100 cm^2 for uranium and its decay products, 200 dpm per 100 cm^2 for thorium and 20 dpm per 100 cm^2 for transuranics and radium-226. The surface contamination levels of Regulatory Guide 1.86 remain conservative compared to the more recently published 2013 ANSI/HPS standard.[6]

[4] Ludlum 3030 Alpha/Beta Counter Specifications. Average of U-238 and Th-230 efficiencies. https://ludlums.com/products/all-products/product/model-3030 (Accessed 10/25/2021)

[5] Although R.G. 1.86 was withdrawn by the NRC in 2016, its acceptable surface contamination levels remain the basis for many current standards for the NRC, DOE, and state regulators. https://www.philrutherford.com/radiation_cleanup_standards.html#RG186 (Accessed 10/25/2021)

[6] ANSI/HPS N13.12 (2013), "Surface and Volumetric Radioactivity Standards for Clearance", May 6, 2013.

Contrary to the statement in the report, the fence line sample set of 11.4 +/- 3.5 CPHr per 4 cm^2 is not *"significantly higher than the background and total sample set results"* (page 4) as the 95% confidence levels overlap each mean. When converted, 13.6 +/- 4.2 dpm per 100 cm^2 is also well below the Reg. Guide 1.86 acceptable removable alpha surface contamination levels quoted above.

Neither the alpha counts per hour in Table 1, nor the beta counts per hour in Table 2 indicate an inverse correlation to distance from SSFL as implied in the text (pages 7, 8).

Beta Count Rate

The conversion factor for beta CPHr per 4 cm^2 to dpm per 100 cm^2 is 1.49 because of the lower beta efficiency of 28%.[7]

The gross removable surface activity measurements (page 5 and Table 2) were

Complete set (n=360)	131 +/- 77 dpm per 100 cm^2
Upwind "background" samples (n=47)	125 +/- 63 dpm per 100 cm^2
SSFL fence line (n=20)	183 +/- 58 dpm per 100 cm^2

The authors again correctly observe that that the three means are not statistically different. The net "background subtracted" complete set mean is 131 - 125 = 6 dpm per 100 cm^2 and the net "background subtracted" fence line mean is 183 - 125 = 58 dpm per 100 cm^2. As with the alpha data, both the gross and net means are significantly less that the Regulatory Guide 1.86 acceptable surface contamination levels for removable beta of 1,000 dpm per 100 cm^2 for general beta/gamma emitters (including Cs-137) and 200 dpm per 100 cm^2 for Sr-90.

Radium-226 Background

Table 3 provides "site-specific" backgrounds obtained from the 2005 DOE Historical Site Assessment (HSA) which utilized the 1998 Bell Canyon dataset for background. It is uncertain why

[7] Ludlum 3030 Alpha/Beta Counter Specifications. Average of Cs-137 and Sr-90 efficiencies. https://ludlums.com/products/all-products/product/model-3030 (Accessed 10/25/2021)

the authors did not use data from the more recent and more comprehensive 2011 EPA Radiological Background Report.[8] The EPA background is added to the revised Table 3 below that includes data in both Bq/kg and pCi/g. Note that the Fairewinds Study uses inconsistent background numbers for radium-226. In the text on page 6, it quotes the radium-226 background as 27 +/- 10.4 Bq/kg, while in Table 3, it uses 25 +/- 11 Bq/kg. Neither of these are correct. The original 2005 DOE HSA Bell Canyon data[9] cited for radium-226 background is 0.94 +/- 0.31 pCi/g which converts to 34.8 +/- 11.5 Bq/kg.

Quality Assurance

In general, for journal papers, one does not have the benefit of reviewing data quality objectives (DQO), quality control (QC) data, calibration data and records, or the radiochemistry laboratory report and narrative. Review of the maximum isotopic data in Table 3 and Table 3 Revised (see below) raises questions that cannot be answered without review of the radiochemistry report.

Although not explicitly stated, it is assumed that the +/- values stated in Tables 1 and 2 are the +/- 1σ (68.3% two-sided confidence level). For the datasets (complete, fence line and background) it is assumed that this is the standard deviation of the data set and represents the variability of the different sample measurements. For the individual samples, this 1σ value is reported by the Ludlum 3030 software and represents the uncertainty of the single measurement.

Isotopic Results

In Table 3 and Table 3 Revised (see below), the +/- values in the background data sets represent the standard deviation (σ) of the dataset and represents the variability of the different sample

[8] EPA, "Final Radiological Background Study Report - Santa Susana Field Laboratory", October 2011. https://www.dtsc-ssfl.com/files/lib_doe_area_iv/bgstudy/Final_SSFL_Radiological_Background_Study_Report.pdf (Accessed 10/25/2021)

[9] DOE, "Historical Site Assessment", Table A-2. May 2005. https://www.etec.energy.gov/Library/Main/SSFLAreaIVHSAVolume%201.pdf#page=75 (Accessed 10/25/2021)

measurements in the background dataset. For the sample set maximum, the +/- 1σ value represents the standard deviation or uncertainty of the single measurement. It should be noted that radiochemistry laboratories conventionally report the uncertainty as +/- 1.96σ not +/- 1σ. However, the paper is silent on this, therefore it is assumed that the authors used a consistent 1σ definition for their +/- values throughout the paper.

It is convention, when reporting radiological laboratory measurements (whether removable surface activity or isotopic concentrations in bulk samples, to provide (1) the measured value, (2) an estimate of the +/- error (1σ or 1.96σ), and (3) the minimum detectable activity[10] (MDA). The MDA is, in general between, 3 to 4 times the σ value. Review of 279 recent isotopic analyses of 58 radionuclides by GEL[11] showed a mean MDA = 3.52σ.

Table 3 Revised below is a revision of the paper's Table 3. Changes made include,

- Correction of the Bell Canyon data for radium-226
- Duplication/conversion of all data to units of picocuries per gram (pCi/g)
- Addition of more recent and relevant EPA background data[12]
- Specification of sample set +/- values as +/- 1σ
- Addition of minimum detectable activity (MDA) column based on MDA = 3.52s
- Addition of qualifier flag column based on comparison of value and MDA to determine if value is non-detect (U)

[10] Minimum detectable activity (MDA) is the a priori activity level that a specific instrument and technique can be expected to detect 95% of the time. When stating the detection capability of an instrument, this value should be used. The MDA is the detection limit, L_D, multiplied by an appropriate conversion factor to give units of activity. MARSSIM. August 2000. https://www.epa.gov/sites/default/files/2017-09/documents/marssim_manual_rev1.pdf#page=639 (Accessed 10/26/2021)

[11] Boeing, "Technical Memorandum - Boeing's Radiological Air Monitoring Data Associated with the Woolsey Fire - Radioisotopic Analyses by GEL Laboratories LLC, December 7, 2018. https://www.boeing.com/resources/boeingdotcom/principles/environment/pdf/Santa_Sue_Boeing_Air_Monitoring_During_Woolsey_Fire.pdf#page=64 (Accessed 10/25/2021)

[12] EPA, "Final Radiological Background Study Report - Santa Susana Field Laboratory", October 2011. https://www.dtsc-ssfl.com/files/lib_doe_area_iv/bgstudy/Final_SSFL_Radiological_Background_Study_Report.pdf (Accessed 10/25/2021)

- Addition of Boeing required MDAs

Comparing the maximums from the sampled dataset to the mean of backgrounds is a statistically inappropriate exercise. More meaningful comparisons could be,

- Parametric comparison of means and variability of the sampled data to the means and variability of background. This determines if the distributions are significantly different. This was done qualitatively for the alpha and beta count rate data.
- Non-parametric comparison of sampled dataset distribution to background distribution, using for example, the Wilcoxon Rank Sum test.
- Compare the maximum of the sampled dataset with a parametric upper bound of the background dataset, such as the 95% upper simultaneous limit (95% USL), the 95/95 upper tolerance limit (95/95 UTL) or a look-up-table (LUT) proposed by EPA.[13]

The authors have done none of these comparisons.

Based on the evaluation of the calculated analysis MDAs, all but U-234 are qualified as U (non-detect). Even the value of U-234 is very close to the MDA.

Inspection of the values of Fairewind's MDAs, shows they are all higher than typical DQO required MDAs that Boeing and its contractors typically require of radiochemistry laboratories to ensure that soil analyses can achieve MDAs at or below background levels (or applicable cleanup standards). Specifically, cesium-137, which was, allegedly, 5 times higher than the mean EPA background, is actually non-detect by a wide margin, and radium-226 which was allegedly 13 times higher than the mean EPA background was also non-detect.

[13] EPA, "Final Technical Memorandum - Look-Up Table Recommendations, Santa Susana Field Laboratory - Area IV Radiological Study", November 27, 2012. https://www.dtsc-ssfl.com/files/lib_doe_area_iv/epaareaivsurvey/techdocs/65778_Final_Tech_Memo_Lookup_Table_Recommendations_112712.pdf (Accessed 10/25/2021)

Comparison to SSFL Area EPA Radiological Data

The 2012 EPA Radiological Survey of Area IV SSFL[14] analyzed 3,735 soil/sediment samples. The major nuclear by-product materials detected above background were,

- 291 (7.8%) samples exceeded cesium-137 background
- 153 (4.1%) samples exceeded strontium-90 background
- 14 (0.4%) samples exceeded plutonium-239 background

The naturally occurring radioactive materials (NORM) detected above background were,

- 1 (0.03%) sample exceeded radium-226 background
- 23 (0.6%) samples exceeded uranium-234 background
- 14 (0.4%) samples exceeded uranium-235 background
- 29 (0.8%) samples exceeded uranium-238 background
- 2 (0.05%) samples exceeded thorium-228 background
- 7 (0.02%) samples exceeded thorium-230 background
- 5 (0.1%) samples exceeded thorium-232 background

The trivially small fraction of EPA soil samples exceeding background, by a factor of 2 or less, for the NORM radionuclides is inconsistent with the focus of the Fairewinds paper on NORM radionuclides especially thorium.

The moderate number of detections of fission products, cesium-137 and strontium-90, above background would suggest that cesium-137 and strontium-90 could be detected offsite following the Woolsey Fire if there had been fire generated transportation of environmental contaminants. Strontium-90 was not reported as detected in the Fairewinds Study. Only one maximum sample with cesium-137 was reported in Table 3 that was shown to be non-detect in Table 3 Revised.

[14] EPA, "EPA Radiological Characterization Study Results", November 2012. https://www.boeing.com/resources/boeingdotcom/principles/environment/pdf/EPA_November_2012_Factsheet.pdf (Accessed 10/26/2021)

It is assumed that the authors were familiar with the EPA Radiological Study of Area IV of SSFL and the relatively moderate prevalence of cesium-137 and strontium-90 compared to NORM radionuclides. The lack of focus on cesium-137 and strontium-90 by the paper's authors suggest that detections, if any, were not prevalent or remarkable

Summary

The isotopic analysis results are questionable, incomplete and appear to have inadequate, undefined DQOs. The MDAs not only exceed the measured values (indicating non-detect results) but also exceed the mean of the background, especially cesium-137 and radium-226, whose MDAs exceed background by a wide margin. Inappropriate comparison is made of isotopic maximums to mean background.

The paper does a great job of demonstrating that radioactive microparticles of thorium monazite are naturally occurring minerals of geologic age. The paper's title attempts to correlate detection of thorium monazite microparticles to the Woolsey Fire. Since thorium monazite originates from geologic age rock, any thorium monazite microparticles in the environment is due to normal weathering of that rock. Thorium monazite microparticles were not created by, the wildfire, the presence of SSFL or its operations.

A significant majority of samples (97% of 360 samples) collected in the study zone registered alpha and beta radioactivity that matched existing area background levels. The maximum alpha and beta count rates were for a sample taken at the maximum distance from SSFL, with one of the highest monazite microparticle content. This suggests that alpha and beta count rates are correlated with monazite content, and not correlated with distance from the site.

The Fairewinds Study results are inconsistent with the results of the 2012 EPA Radiological Study of Area IV of SSFL.

It is hard to imagine how this paper passed peer review.

Table 3 Revised
Site-specific Background Activities vs. SSFL Sample Maximums

Isotope	1998 Bell Canyon Bkgd Bq/kg		2011 EPA Bkgd Bq/kg		Sample Set Maximum Bq/kg			Qualifier Flag[e]	Sample ID	Boeing Required MDA (Bq/kg)
	Mean	+/- 1σ	Mean	+/- 1σ	Value	+/- 1σ[c]	MDA[d]			
Cs-137	1.96	1.85	3.3	1.2	16.7	11.8	41.5	U	161 S	1.85
U-234	27	7.8	41.8	8.3	91	25	88.0		126 A	37
U-235	1.63	0.74	2.3	0.7	4.1	3.3	11.6	U	122 S	3.7
U-238	27	7.8	42.1	8.4	72.5	21.8	76.7	U	126 A	37
Th-228	32.2	9.2	78.9	17.1	82.9	27	95.0	U	126 A	37
Th-230	27	10.4	45.7	9.1	164	49	172	U	148 S	37
Th-232	32	9.2	59.6	14.8	74.4	24.8	87.3	U	126 A	37
Ra-226[b]	34.8	11.5	50.4	11.5	648	307	1081	U	021 S	37

Isotope	1998 Bell Canyon Bkgd pCi/g[a]		2011 EPA Bkgd pCi/g[a]		Sample Set Maximum pCi/g[a]			Qualifier Flag[e]	Sample ID	Boeing Required MDA (pCi/g)
	Mean	+/- 1σ	Mean	+/- 1σ	Value	+/- 1σ[c]	MDA[d]			
Cs-137	0.053	0.05	0.0897	0.0319	0.451	0.319	1.12	U	161 S	0.05
U-234	0.73	0.21	1.129	0.223	2.46	0.676	2.38		126 A	1.0
U-235	0.044	0.02	0.0634	0.020	0.111	0.089	0.31	U	122 S	0.1
U-238	0.73	0.21	1.137	0.227	1.96	0.589	2.07	U	126 A	1.0
Th-228	0.87	0.25	2.132	0.461	2.24	0.730	2.57	U	126 A	1.0
Th-230	0.73	0.28	1.234	0.247	4.43	1.32	4.66	U	148 S	1.0
Th-232	0.86	0.25	1.611	0.400	2.01	0.670	2.36	U	126 A	1.0
Ra-226[b]	0.94	0.31	1.361	0.311	17.51	8.30	29.2	U	021 S	1.0

[a] 1 pCi/g = 37 Bq/kg

[b] Bell Canyon Ra-226 is 0.94 +/- 0.31 pCi/g (34.8 +/- 11.5 Bq/kg), not 27 +/- 10.4 Bq/kg or 25 +/- 11 Bq/kg

[c] Sample set maximum errors are assumed to be 1σ, since the paper is silent on this question

[d] Average MDA from 279 GEL analyses of 58 radionuclides = 3.52σ

[e] If value is < MDA then the radiochemistry laboratory will flag the result as U (non-detect)

Appendix

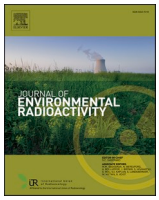
Radioactive particles related to the Woolsey Fire in Simi Valley, CA

Journal of Environmental Radioactivity 240 (2021) 106755

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad



Radioactive microparticles related to the Woolsey Fire in Simi Valley, CA

Marco Kaltofen [a,*], Maggie Gundersen [b], Arnie Gundersen [b]

[a] Worcester Polytechnic Institute, Dept. of Physics, 100 Institute Road, Olin Hall, Worcester, MA, 01609, USA
[b] Fairewinds Energy Education, 520 Folly Road, Ste. 25-344, Charleston, SC, 29412, USA

A B S T R A C T

In November 2018, the Woolsey Fire burned north of Los Angeles, CA, USA, potentially remobilizing radioactive contaminants at the former Santa Susana Field Laboratory, a shuttered nuclear research facility contaminated by chemical and radiochemical releases. Wildfire in radiologically contaminated zones is a global concern; contaminated areas around Chernobyl, Fukushima, Los Alamos, and the Nevada Nuclear Test Site have all experienced wildfires. Three weeks after the Woolsey Fire was controlled, sampling of dusts, ashes, and surface soils (n = 360) began and were analyzed by alpha- and beta-radiation counting. Samples were collected up to a 16 km radius from the perimeter of the laboratory. Controls and samples with activities 1σ greater than background were also examined by alpha and/or gamma spectroscopy or Scanning Electron Microscopy with Energy Dispersive X-ray analysis. Of the 360 samples collected, 97% showed activities at or close to site-specific background levels. However, offsite samples collected in publicly-accessible areas nearest to the SSFL site perimeter had the highest alpha-emitting radionuclides radium, thorium, and uranium activities, indicating site-related radioactive material has escaped the confines of the laboratory. In two geographically-separated locations, one as far away as 15 km, radioactive microparticles containing percent-concentrations of thorium were detected in ashes and dusts that were likely related to deposition from the Woolsey fire. These offsite radioactive microparticles were colocated with alpha and beta activity maxima. Data did not support a finding of widespread deposition of radioactive particles. However, two radioactive deposition hotspots and significant offsite contamination were detected near the site perimeter.

1. Introduction

The Santa Susana Field Laboratory (SSFL), a former center for nuclear research, is located in the Simi Hills area of Ventura County, California, USA. The Simi Hills are a low rocky mountain range. SSFL was developed during the late 1940s and 1950s on a 1152-ha site approximately 48 km northeast of Los Angeles in what was then a rural area. The research site hosted rocket engine tests, conducting 17,000 engine tests during the 60-year life of the site (Boeing 2021). The site hosted multiple atomic reactors for nuclear research under a U.S. Department of Energy (DOE) license. One of the site's test reactors, the Sodium Reactor Experiment, experienced a partial meltdown and vented an unmonitored amount of radiation into the environment in July 1959 (US DOE 2020a). Other unmonitored documented radiation releases from other test reactors and burn pits occurred, ending in 1988 when all nuclear research at the site was terminated (EPA 2012a).

Since the end of SSFL operations, studies by the US Environmental Protection Agency (EPA) have detected SSFL-specific radioactive contaminants, including cesium-137, cobalt-60, radium-226, and technetium-99. EPA investigators have also detected the more commonly found isotopes of uranium and thorium and their daughters above naturally-occurring background concentrations. These investigations confirmed that some radioactive contaminants from SSFL had migrated to areas outside the site perimeter (EPA 2012b; EPA 2012c). Uranium and thorium and their decay products have alpha- and beta-emitting isotopes and may be naturally-occurring or of industrial origin (ATSDR 2013; Kaltofen 2019).

During the three decades following the 1988 closure of operations at the SSFL site, the Los Angeles suburbs have encroached on the Simi Hills's once-rural land. Changes in regional and global climate favoring increased drought along with actual weather conditions on the day of the fire start meant that "The conditions for a large-scale fire incident were perfect" (LA County 2019). Today, 500,000 people live within 16 km of the SSFL site. In early November 2018, a fire began on the SSFL site (US DOE 2020b). This fire, which became known as the Woolsey Fire, was spread by winds gusting more than 70 km per hour and rapidly engulfed 39,500 ha, devoured 1500 structures, killed three people, and spread as far as coastal California at Malibu. The fire spread as much as 25 km SW of SSFL, which is a formidable spread. In addition, an area adjacent to the SSFL was the origin of the wildfire. Financial losses exceeded $1.5 Billion (LA Times, 2018).

"Wildfires typically mobilize lead-210, bismuth-210, and polonium-210," all of which are decay products of naturally-occurring radon (Los Alamos 2000). In addition, wildfire, and climate change-driven wildfire

* Corresponding author. ,
E-mail address: mpkaltofen@gmail.com (M. Kaltofen).

https://doi.org/10.1016/j.jenvrad.2021.106755
Received 25 May 2021; Received in revised form 13 August 2021; Accepted 27 September 2021
Available online 8 October 2021
0265-931X/

in particular, has been a driver of radioactive contaminant transport in forest fires at Chernobyl in Ukraine (Evangeliou 2015) and the Cerro Grande Fire at Los Alamos that transported cesium-137 and strontium-90-contaminated materials (Los Alamos 2001). The potential impact of climate change on radionuclide transport is two-fold, as increasing drought, fire frequency, and increased precipitation intensity result in greater post-fire stormwater transport of contaminated sediments. This increase is over and above the typical increase in runoff and sediment yield after wildfires (ibid). At the Hanford Nuclear Reservation in Washington State, the Wautoma wildfire detected elevated gross alpha and gross beta activities in fire plume air samples not attributable to natural radon (WADOH 2008). Multiple wildfires have ignited in the restricted zones surrounding the Fukushima Dai-ichi Nuclear Power Station in Japan. An April 2017 forest fire in a radioactively-contaminated part of Namie, Fukushima Prefecture, Japan, was extinguished using fire suppressing helicopters (Mainichi 2017). The fires resulted in a localized increase of cesium-137 activities of 3–9 times at distances of up to 8 km from the wildfire (Safecast 2017). In May 2021, the Cherrywood Wildfire at the Nevada (USA) National Security Site approached the portion of the site impacted by surface nuclear material contamination, requiring plans to evacuate firefighters (PVT citing BLM 2021).

This study's objective was to determine if a low-cost crowdsourced sampling method could be rapidly initiated to establish whether SSFL-specific contaminants had migrated beyond the site boundary into the surrounding community because of Woolsey Fire-related ash and dust transport. This study used community-volunteer Citizen-Science methodology previously developed worldwide and applied at other radiologically contaminated nuclear power plant and nuclear waste sites and communities surrounding them. Citizen-Science is not a new phenomenon; it began more than 2000 years ago in ancient China to monitor and track migratory locusts that destroyed harvests (Nature 2018).

Worldwide, Citizen-Science is now a crucial scientific method that enables real-time scientific data to move beyond corporate scientists and into the hands of those most impacted who must design and implement environmental and health mitigation projects (IAEA 2016). Moreover, Citizen-Science work is open-source. As a result, locally impacted communities, states, provinces, prefectures, and national governments may realize the magnitude of ecological and public health impacts of chemical, radioactive, and other human-made detritus to arrange remediation procedures and public financing (Silverton 2011).

There has been an uptick in the use of Citizen-Science during the last decade due to the wide availability of internet access and accurate, inexpensive GPS (Global Positioning Systems). It is a low-cost solution that allows large-scale sampling despite limited funding for scientific and health studies. Citizen-Science fosters international cooperation and enables scientists to get timely answers that would have been unaffordable with any other methodology, as exhibited in the analysis of air quality Citizen-Science in Antwerp Belgium. This Flemish Environmental Protection Agency and Citizen-Science partnership included a team of 50,000 non-academic volunteers (Nature 2018).

In the United States, federal statutes note that "crowdsourcing and Citizen-Science projects have additional unique benefits, including accelerating scientific research, increasing cost-effectiveness to maximize the return on taxpayer dollars, addressing societal needs, providing hands-on learning in STEM, and connecting members of the public directly to Federal science agency missions and each other; " (USC 2021). Citizen-Science has been instrumental, for example, in studies of the Fukushima Dai-ichi region and at control sites in the U.S. (Kaltofen 2018).

Typically radiological migration studies look at gross measures of radioactivity in bulk materials such as soils and sediments. However, this study also included data on microparticles of dust or soil. These particles contain sufficiently-high concentrations of radioactive elements to exert an internal radiation dose if the microparticle is inhaled via radioactive dust particles suspended in the air or incidentally ingested from contact with soil or dusts containing radioactive microparticles. Once radioactive microparticles enter the body by either pathway, the body will continue to be exposed to radiation until the material is excreted via urine or feces (Japan MOE 2021). The radioactive isotopes detected in bulk samples and microparticles are alpha-emitters, including primarily uranium-234, 235, and 238; thorium-228, 230, and 232; and radium-226 and 228; as well as beta-emitters, of which cesium-137 was the most prevalent beta-nuclide found at the SSFL site (DOE 2005).

These alpha-nuclides occur both naturally and industrially. For example, thorium occurs naturally as thorium-232, a primordial radionuclide. Typically this radioactive element is found as monazite, a crystalline mineral composed of cerium, lanthanum, rare earth elements, calcium, phosphorous, plus thorium, and occasionally also uranium (Kaltofen 2018). Thorium also occurs as a decay daughter of uranium-234 and 238. Fertile thorium-232 is also used as a refined metal in nuclear materials and in industrially-produced monazite-type materials as a nuclear waste form (Ibid). This study uses the activity, location, and elemental composition of particulate matter that contains uranium, radium, and thorium to distinguish between natural and industrial alpha-emitters.

The large number of samples collected was possible given the involvement of Citizen-Science volunteers. Given the low percentage of dust samples ultimately found to contain significant activity, citizen-scientists were essential to getting a sufficiently large number of samples from which to draw conclusions regarding this site.

2. Methods

A total of 360 samples of house dust, surficial soils, and ash were collected for this study. Sample collection began in December 2018 (less than one month after the Woolsey Fire began) and ended by February 2019. Samples were collected from private residential properties (with property owners' permissions), public areas, and public areas directly adjacent to the SSFL site fenceline. The samples were collected using community-volunteers from the Los Angeles area. These community-volunteer citizen-scientists had received training in sample collection and safety protocols before collecting samples. The locus of areas sampled extended to a radius of approximately 16 km around the perimeter of the SSFL-site. This area encompasses rural, urban, suburban, and undeveloped mountainous locations.

The samples collected came from the Woolsey Fire zone (Fig. 1) or unaffected areas generally easterly and upwind of the fire zone. Fig. 2 shows the Woolsey Fire burn area overlaid with the prevailing Santa Ana winds. This represents wind direction and speed for the fire period (NASA 2018).

Each sample was assigned a unique identifier and tagged with latitude and longitude data acquired using a handheld Global Positioning System (GPS). No radiation monitoring devices were employed to bias sample collection. One sampling team used a b-Geigie® rate counter with an integrated data-logger to produce a record of gamma readings and GPS coordinates during the SSFL site perimeter sampling. These data were not reviewed until after the completion of sample collections. All bulk samples (n = 124 of 360) were collected in double Ziplock® bags with GPS and identification data affixed to both bags. Depositional dust and ash samples were collected from residential locations using Zefon® Biotape® microscope slides with a 4-cm^2 adhesive area that collected a thin layer of dust, soil particles, or ashes (n = 236 of 360). Lift-tape samples were prepared by contacting the sampled surface with the adhesive portion of the Bio-Tape slide, removing it, and then returning the slide to its individual case (Barbieri, 2009). Samples were assigned a suffix (A, D & S) for ashes, surface dusts, and soils, respectively.

When packed for shipping, samples were placed inside two boxes with a gap between the boxes to create a geometry that reduced the potential for any radiation exposure during transit. Prior to final

Fig. 1. Map of the sampled area showing Woolsey Fire (2018) extent and SSFL (DTSC, 2020) (low-res version, actual is high-res).

shipping, all samples were prescreened using an International Medcom Inspector rate counter that could detect alpha, beta, and gamma activity (using a halogen-quenched Geiger-Mueller tube with an effective diameter of 45 mm and a mica window density of 1.5–2.0 mg/cm^2).

All samples were analyzed using the same geometry by preparing lift-tape (Bio-Tape®) samples. The samples were collected directly onto lift-tapes in the field or as a monolayer from the air-dried and homogenized bulk samples in the laboratory. There was no sieving or other type of particle size adjustment during preparation. Slides were counted for three 1-h counts using a Ludlum Model 3030 two-channel alpha-beta rate counter. Bulk samples with alpha and beta counts greater than 1σ above the respective means were also analyzed by alpha and gamma spectroscopy and/or Scanning Electron Microscopy/Energy Dispersive X-ray Spectroscopy analysis (SEM/EDS).

On selected samples, alpha spectroscopy (with pretreatment by digestion and chemical separation of approximately 1.0 g samples, followed by planchet counting for radium, thorium, and uranium alpha decays) was performed by Eberline Analytical Corp, a commercial testing laboratory in Oak Ridge, TN. Eberline also performed a high-resolution gamma analysis using quantitative high-purity Germanium [HPGe] spectroscopy (for cesium-137, cesium-134, and uranium/thorium decay products using air-dried bulk samples).

SEM/EDS analysis was performed by Microvision® Laboratories of Chelmsford, MA, using a Bruker® X-Flash® Peltier-cooled silicon drift detector (SDD). The electron beam current was 0.6nAmperes, accelerated to a voltage of 0.5–60 keV. Air-dried Biotape® slides were carbon-coated before X-ray analyses. A Robinson Detector was used to scan for high-Z materials among the particles. The SDD detector can quantify the elemental composition of particles with an approximate limit of detection of 0.2%. SEM/EDS does not detect radioactive decays but will efficiently detect high-Z elements such as uranium and thorium that contribute to the alpha activity. Prior to the SEM analysis, slides were weighed, preloaded, and post-loaded, and were found to contain (on average) 4.4 ± 2.1 mg of the material, based on the mass gain over the tare value for the Bio-Tape slides (Kaltofen 2020).

3. Results

3.1. Alpha count rate

The complete set of study samples had a mean net alpha count rate of 7.6 ± 3.0 counts per hour (CPHr) for each 2 cm × 2 cm slide (α CPHr 4 cm^{-2}). The mean of background samples (soils/dusts east and upwind of SSFL and the fire zone) was 7.4 ± 4.4 CPHr 4 cm^{-2}. The two means are not significantly different, attesting to the scattered nature of the elevated activity samples. One outlier was neglected in calculating σ for the mean. The collection points of samples with notable alpha count rates and other detections are shown in Fig. 3. Samples with alpha count rates that were about 2σ greater than the mean are shown in Table 1. Only seventeen of the 360 samples collected had alpha count rates about 2σ greater than the mean, with σ based on counting variability, not assuming the actual count data were normally distributed (Figs. 1–6).

Of the 360 samples collected, twenty (20) were sampled from the perimeter (fenceline) of the SSFL. The mean alpha count rate for the

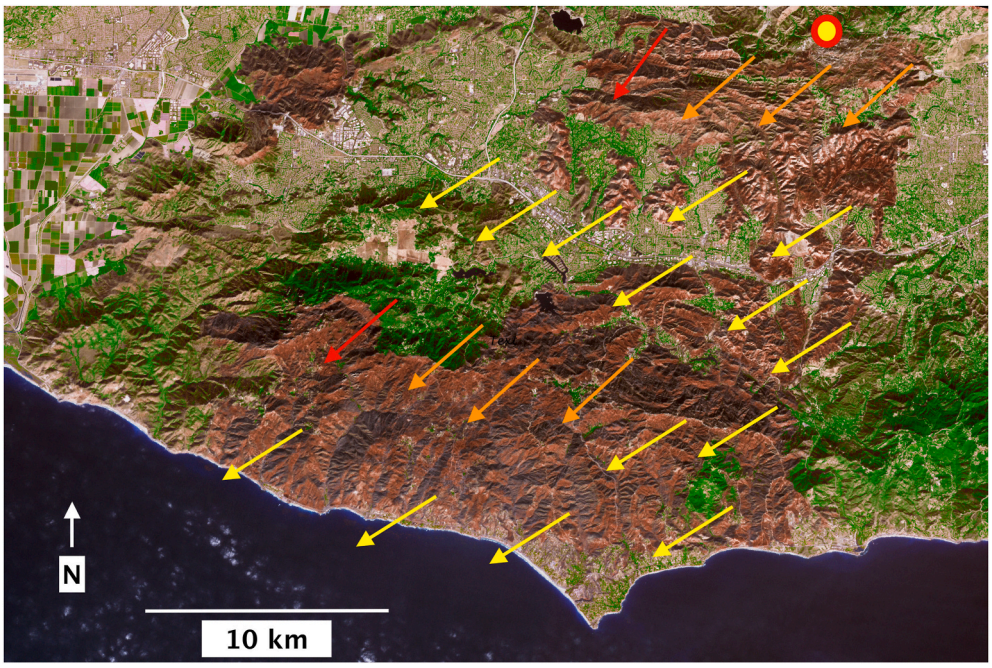


Fig. 2. Map of Woolsey Fire extent with wind pattern overlay (NASA, 2018) showing approximate fire origin (circle), wind direction, and maximum wind gust speeds (yellow 30 mph, orange 30-40 mph, red >40 mph). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

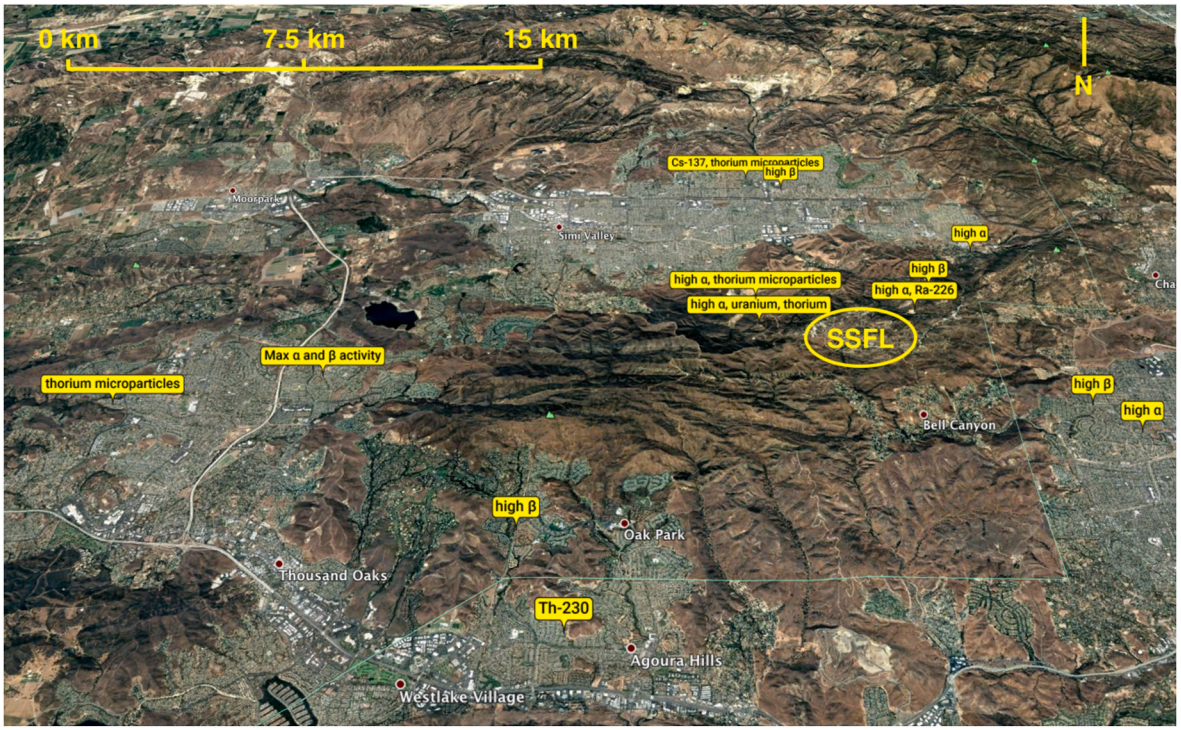


Fig. 3. Map of the notable alpha, beta, gamma, and SEM/EDS results, adapted from Google Earth (2020) (low-res version, actual is high-res).

fenceline sample set was 11.4 ± 3.5 CPHr 4 cm^{-2}. This mean is significantly higher than the means for the background and total sample set results.

A deposited ash sample (180 A) collected 15 km from the SSFL perimeter (in Thousand Oaks, CA) had the highest alpha count rate of the entire sample set. X-ray microanalysis (described later in the SEM/EDS results) detected multiple thorium-bearing radioactive microparticles in this same ash sample. Thus, the data for sample 180 A is consistent with fire-transported radioactive microparticles. In Table 1, samples taken nearest the SSFL fenceline had generally higher alpha count rates and were therefore over-represented compared to the rest of the sample set.

Table 1
Net Alpha counts per hour (α-CPHr) over instrument background [ID identifier: A = Ash, D = Dust, S=Soil, Control = pre1945 wood samples].

ID	α-CPHr	km to SSFL	direction to SSFL
180 A	140.4 ± 15.1	15 km	SSW
020 S	23.1 ± 2.1	<0.5 km[a]	N
150 S	20.7 ± 2.5	<0.5 km[a]	N
332D	20.4 ± 1.0	4.4 km	SE
126 A	20.1 ± 3.8	<0.5 km[a]	W
129 S	19.7 ± 6.4	<0.5 km[a]	W
169 S	18.4 ± 4.4	11.6 km	ESE
161 S	17.4 ± 3.1	5.5 km	N
218D	16.4 ± 1.0	12.7 km	W
206D	16.4 ± 4.0	15 km	W
025 S	15.7 ± 5.5	<0.5 km[a]	N
170 S	15.4 ± 2.1	1.7 km	NW
148 S	15.4 ± 3.8	9.5 km	SW
021 S	15.4 ± 6.1	<0.5 km[a]	N
024 S	15.1 ± 4.5	<0.5 km[a]	N
122 S	15.1 ± 6.4	6.1 km	NW
All samples	7.6 ± 3.0	–	–
Fenceline only	11.4 ± 3.5	–	–
Background (S)	7.4 ± 4.4	varies	E
Control	0.5 ± 1.0	NA	NA
Control (ashed)	4.2 ± 3.1	NA	NA

[a] Fenceline sample adjacent to SSFL.

3.2. Beta count rate

The complete set of study samples had a mean net beta count rate of 87.7 ± 51.8 CPHr for each 2 cm × 2 cm slide (CPHr 4 cm^{-2}). The instrument background net beta count rate was 0.0 ± 79 CPHr 4 cm^{-2}. The mean of background samples (n = 47, soils/dusts east and upwind of SSFL and the fire zone) was 84 ± 42 CPHr 4 cm^{-2}. For the twenty (20) samples from the perimeter (fenceline) of the SSFL, the mean beta count rate was 123 ± 39 CPHr 4 cm^{-2}. The three means are not significantly different, further confirming the elevated activity samples' isolated and geographically-separated nature. This result may also reflect that of the beta-emitting radioisotopes found at SSFL itself, only one, cesium-137, exists at significant levels above background. None of the radioactive microparticles detected in the study included any form (radioactive or stable) of cesium. This result is consistent with the finding that beta counts for the fenceline set were, on average, within 1σ of the whole set (see Table 2).

The beta count rate geographical distribution was similar to that for alpha count rates, with the same maximally-elevated results for sample 180 A (the Thousand Oaks, CA ash sample) and a preponderance of SSFL fenceline samples in this elevated count rate group. Some degree of correspondence between alpha and beta results is anticipated given the radioactive decay chains for thorium, uranium, and radium. These are identified SSFL contaminants of concern (EPA 2012b; EPA 2012c). All yield both alpha and beta decays in their respective decay chains (ATSDR 2013). Many higher beta count rate samples cluster to the west and southwest of the Santa Susana Field Lab (SSFL) in an area within the fire zone. Excluding fenceline samples, only 3 of 22 samples in the high-count rate group are located in the eastern and generally upwind non-fire zone. While these findings mirror the location fire zone, it also reflects the greater sampling density in the fire zone compared to areas located to the east of SSFL (n = 305 fire zone or downwind vs. n = 55 upwind, non-fire zone and easterly).

3.3. Isotopic data

As with the alpha and beta count data, the highest isotopic activities were found in samples collected nearest the SSFL site, several additional scattered detections above the background levels in the fire-impacted areas. The highest activities for sampled uranium and thorium isotopes originated from an ash sample collected near the western edge of Santa Susana Field Lab, indicating that the Woolsey fire produced ashes contaminated by radioactivity, whether natural or industrial, from the material at SSFL. Table 3 summarizes the highlights of the alpha and gamma spectroscopic analyses. This study's maximum sample activity of radium-226, 648 ± 307 Bq kg^{-1}, was more than 18 times the DOE

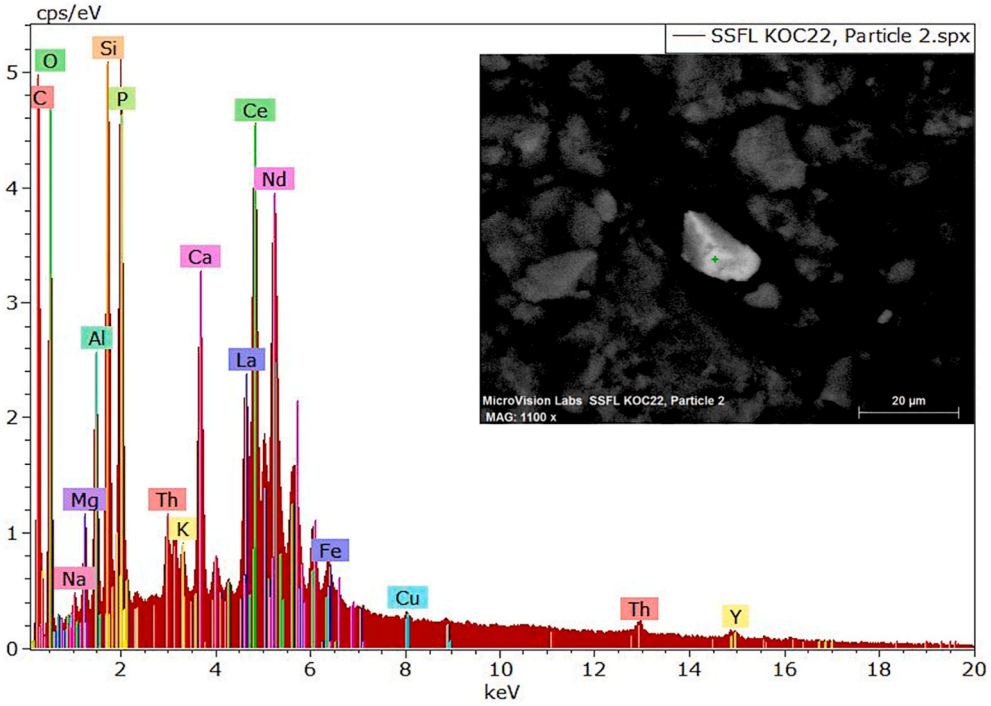


Fig. 4. SEM/EDS Data Particle SSFL22S.2 with 19-µm long axis diameter and with elemental composition of 2.1% Th, 11.6% Ce, 6.2% P, 5.8% La, 5.1% Nd, and 3.1% Ca. The sample was collected at the SSFL fenceline.

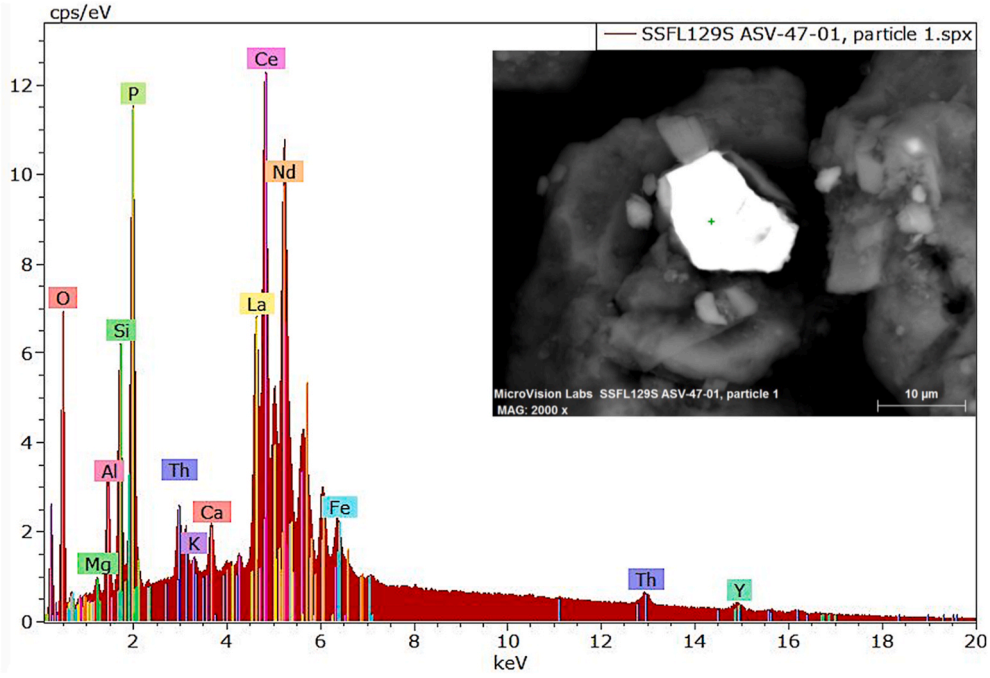


Fig. 5. SEM/EDS Data Particle SSFL129S.1 with 16-µm long axis diameter and with elemental composition of 5.0% Th, 29.9% Ce, 16.6% La, 15.3% P, 12.6% Nd, and 1.2% Ca. The sample was collected at the SSFL fenceline.

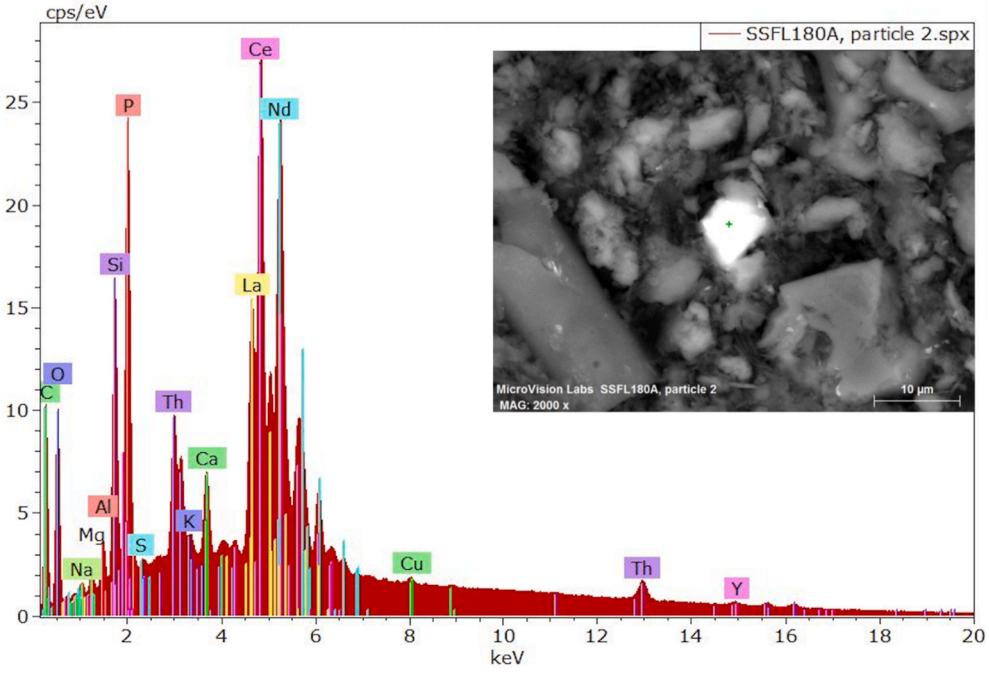


Fig. 6. SEM/EDS Data Particle SSFL180A.2 with 8-µm long axis diameter and with elemental composition of 9.6% Th, 29.8% Ce, 15.2% La, 15.0% P, 10.9% Nd, and 1.9% Ca. This ash sample was collected 15 km SSW of the SSFL in Thousand Oaks, CA. This ash sample also had the maximum alpha and beta count rates of the entire 360-sample set.

reported background activity, 27 ± 10.4 Bq kg^{-1} (DOE 2005). This surface soil sample was collected just outside the fenced perimeter north of SSFL. Therefore, radium-226 is an SSFL-specific contaminant of concern (EPA 2012c).

Similarly, the highest activities for uranium-234, uranium-238, thorium-228, and thorium-232 were all found in the ash sample collected at the west-side fence perimeter immediately outside of SSFL. The elevated cesium-137 activity of sample 161 S was eight times background and came from Simi Valley, CA. This same 161 S sample also contained thorium monazite particles. The elevated thorium-230 activity of six times background was found in a sample collected in Agoura Hills, CA.

3.4. SEM/EDS results

Of the thirteen radioactive microparticles detected, nine were in the

Table 2
Net Beta in counts per hour (β-CPHr) over instrument background [ID identifier: A = Ash, D = Dust, S=Soil, Control = pre1945 wood samples].

ID	β-CPHr	km to SSFL	direction
180 A	322 ± 21	15 km	SSW
012	221 ± 38	<0.5 km[a]	N
023	188 ± 49	<0.5 km[a]	N
020	185 ± 32	<0.5 km[a]	N
317D	184 ± 16	8 km	SW
021	182 ± 78	<0.5 km[a]	N
171 S	179 ± 79	3 km	N
210D	175 ± 21	11 km	SSW
220D	175 ± 28	5.6 km	WNW
289D	168 ± 15	10 km	SE
170 S	168 ± 98	1.7 km	NW
182D	167 ± 23	7 km	SSE
168 S	165 ± 13	4.6 km	NW
311D	164 ± 16	12.7 km	SW
259D	164 ± 19	4.1 km	NW
110 S	163 ± 8	16 km	NW
108 S	162 ± 56	14 km	NW
173 S	157 ± 28	10.6 km	SW
164 S	156 ± 23	5.4 km	S
109 S	153 ± 39	14.5 km	NW
176 S	152 ± 13	5.5 km	SW
239D	152 ± 45	4.1 km	E
134 S	151 ± 24	3.3 km	NNW
197D	151 ± 27	8.3 km	NNW
231D	150 ± 65	12.5 km	SW
161 S	149 ± 19	0.5 km	W
All samples	96 ± 45	–	–
Fenceline only	123 ± 49	–	–
Background	84 ± 42	varies	E
Control	13 ± 53	NA	NA
Control (ashed)	115 ± 39	NA	NA

[a] Fenceline sample adjacent to SSFL.

Table 3
Site-specific background activities (US DOE, 2005) vs. SSFL sample maximums. Data are in Becquerels per kilogram (Bq kg-1). Sample IDs and matrices are also shown.

Isotope	Background	Sample set max.		Note	Location
Cs-137	1.96 ± 1.85	16.7 ± 11.8	161 S	high β	Simi Valley
U-234	27. ± 7.8	91 ± 25	126 A ash	high α	<0.5 km (W)
U-235	1.63 ± 0.74	4.1 ± 3.3	122 S	high α	Simi Valley
U-238	27 ± 7.8	72.5 ± 21.8	126 A ash	high α	<0.5 km (W)
Th-228	32.2 ± 9.2	82.9 ± 27	126 A ash	high α	<0.5 km (W)
Th-230	27 ± 10.4	164 ± 49	148 S	high α	Agoura Hills
Th-232	32 ± 9.2	74.4 ± 24.8	126 A ash	high α	<0.5 km (W)
Ra-226	25 ± 11	648 ± 307	021 S	high α & β	<0.5 km (N)

fence line area (<0.5 km from SSFL), four were found 5.5 km north of SSFL in Simi Valley, or 15 km west of SSFL in Thousand Oaks. The range of thorium compositions in detected monazite particles was 0.8–11.2 percent thorium by mass, with a mean of 5.3% ± 3.9% (Table 4). Thorium was used extensively at SSFL, including in the Sodium Reactor Experiment Complex, where the reactor used 93% uranium-thorium metal alloy fuel (EPA 2012a).

All thirteen radioactive microparticles contained percentage thorium concentrations as thorium monazite (Figs. 4–6). Monazite is a naturally occurring radioactive calcium-phosphate mineral containing 3%–11% thorium, rare earth elements including La, Ce, Pr, Nd, Sm, Eu, and Gd; and (typically) less than 1% uranium (Tew 1968; Hayaton 2019). Thorium monazite may also originate from industrial wastes. This mineral is a source of thorium and uranium for the nuclear industry, a byproduct of metal refining, and monazite has also been used experimentally as a waste storage form for actinides (Boatner 1980; Paschoa 2010; Ewing 1999).

Table 4
Radioactive microparticles detected [ID identifier: A = Ash, D = Dust, S=Soil].

Sample ID	Particle composition	Location	
206D	11.2%	Th as monazite	Thousand Oaks, CA
180 A	9.8%	Th as monazite with Lead (Pb)	Thousand Oaks, CA
161 S	2.7%	Th as monazite	Simi Valley, CA
126 A[a]	9.8%	Th as monazite	
126 A[a]	9.0%	Th as monazite	
129 S[a]	5.0%	Th as monazite	
129 S[a]	4.4%	Th as monazite	
129 S[a]	2.2%	Th as monazite	
021 S[a]	2.1%	Th as monazite	
021 S[a]	0.9%	Th as monazite	
021 S[a]	0.8%	Th as monazite	
022 S[a]	1.4%	Th as monazite	
022 S[a]	0.9%	Th as monazite	

[a] Fenceline sample adjacent to SSFL.

The radioactive microparticles detected in this study are not definitively natural or industrial. Notably, however, the most significant number of monazite particles were detected at the SSFL fenceline, evidencing that, whether industrial or natural, the Santa Susana Field Laboratory or its exposed bedrock is a potential source of the thorium monazite particles detected in this study. Also notable, the fire ash sample 180 A from Thousand Oaks, CA that had the highest alpha and beta count rates of all 360 samples and a 9.8% thorium particle, also contained particles composed primarily of lead, an industrial metal. This finding suggests that the ash in this sample resulted from a manufactured substrate's combustion or an industrially contaminated material. Finally, as previously noted, ash samples collected at the SSFL site's perimeter contained thorium isotopes at up to 500% of the site-specific background creating further evidence that thorium mobilizes from the Santa Susana Field Laboratory.

California Environmental Protection Agency (CalEPA) data found a similar pattern of scattered localized areas of elevated radioactivity samples. CalEPA found elevated alpha and beta activity at a Bell Canyon location 1.4 km S of the SSFL. Further analysis by Lawrence Livermore National Laboratory identified isotopes consistent with low levels of cesium-137 at one site and alpha emitters [such as radium or thorium] in the Bell Canyon sample (DTSC 2020). These CalEPA findings mirror those of this study, with generally unremarkable data in the fire zone save for the scattered higher-activity locations where ashes were deposited. Likewise, US EPA data has found thorium levels significantly above site background activities but has not used any X-ray techniques to determine the excess thorium's chemical or mineral state (EPA 2012c). Based on this study's findings, any future proposed environmental analyses should include an SEM/EDS analysis of onsite dusts, remnant ashes, and surface soils to determine if similar thorium monazite particles exist onsite beyond the detection rate found for fenceline and offsite samples.

4. Conclusions

A significant majority of samples (97% of 360 samples) collected in the study zone registered radioactivity levels that matched existing area background levels. Nevertheless, some ashes and dusts collected from the Woolsey Fire zone in the fire's immediate aftermath contained high activities of radioactive isotopes associated with the Santa Susana Field Laboratory (SSFL). The data show that Woolsey Fire ash did, in fact, spread SSFL-related radioactive microparticles, and the impacts were confined to areas closest to SSFL and at least three other scattered locations in the greater Simi Valley area. Alpha and beta counting, high-

resolution alpha and gamma spectroscopy, and X-ray microanalysis using SEM/EDS confirmed the presence of radioactive microparticles in the Woolsey Fire-related ashes and dusts.

Most of the fire-impacted samples found near the SSFL site's perimeter were on lands accessible to the public. There were, however, scattered localized areas of increased radioactivity due to the presence of radioactive microparticles in ash and recently-settled dusts collected just after the Woolsey fire. These radioactive outliers were found in Thousand Oaks, CA, and Simi Valley, CA, about 15 and 5 km distant from SSFL, respectively. The Thousand Oaks samples had alpha count rates up to 19 times background, and X-ray spectroscopy (SEM) identified alpha-emitting thorium as the source of this excess radioactivity. Excessive alpha radiation in small particles is of particular interest because of the relatively high risk of inhalation-related long-term biological damage from internal alpha emitters compared to external radiation.

The nuclides identified as the sources of excess radioactivity in impacted samples were predominately isotopes of radium, uranium, and thorium. These have naturally-occurring sources, but these isotopes are also contaminants of concern at SSFL and were detected at generally increasing activities as the distance from SSFL decreased. In addition, the number of radioactive microparticles per gram of particulate matter also increased strongly with decreasing distance from SSFL. These data demonstrate that fire and/or other processes have spread SSFL contamination beyond the facility boundary.

Credit author statement

Marco Kaltofen, conceptualization, methodology, formal analysis, writing – original draft; M. Gundersen, Resources, data curation, editing, Project Administration, funding acquisition; A. Gundersen, Investigation, editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Funding is provided by the Samuel Lawrence Foundation, the Davis-Dauray Family Fund, and Fairewinds Energy Education. In addition, this study received nonfinancial support for sample collection from the Physicians for Social Responsibility-Los Angeles chapter (PSR-LA) and Kerry Anne O'Connor. Additional nonfinancial support was received from the Natick Community Organic Farm of Natick, Massachusetts (USA), which provided samples of intact and ashed wooden beams from its 1815 barn for use as controls.

References

ATSDR, 2013. Agency for Toxic Substances and Disease Registry, Division of Toxicology and Human Health Sciences, TP150 Toxicological Profile for Uranium. February 2013.

Barbeiri, A., 2009. Wipe sampling for characterization of non-compactable radioactive waste. In: 2009 International Nuclear Atlantic Conference - INAC 2009 Rio de Janeiro, RJ, Brazil, September 27 to October 2, 2009. ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN, ISBN 978-85-99141-03-8.

Boatner, L.A., Beall, G.W., Abraham, M.M., Finch, C.B., Huray, P.G., Rappaz, M., 1980. Monazite and other lanthanide orthophosphates as alternate actinide waste forms. In: Northrup, C.J.M. (Ed.), Scientific Basis for Nuclear Waste Management. Advances in Nuclear Science & Technology. Springer, Boston, MA. https://doi.org/10.1007/978-1-4684-3839-0_35.

Boeing, 2021. Santa susana's extraordinary past. URL accessed 5/5/2021. https://www.boeing.com/principles/environment/santa-susana/extraordinary-past.page.

DTSC, 2020. Final Summary Report of Woolsey Fire, Impacts of SSFL & Surrounding Communities Sampling Results. California Environmental Protection Agency, Department of Toxic Substances Control. December 2020.

EPA 2012a, URL (accessed 11/19/20) https://www.etec.energy.gov/Library/Cleanup_and_Characterization/Soil/Co-Located/1-Final%20FSP%20for%20Soil%20Sampling%20030512.pdf.

EPA, 2012b. HydroGeologic, inc. In: Final Gamma Radiation Scanning Report Area Iv Radiological Study Santa Susana Field Laboratory Ventura County. California, EPA Contract Number: EP-S7-05-05 Task Order Number: 0038, Oct. 17, 2012.

EPA, 2012c. HydroGeologic, inc. In: Final Radiological Characterization of Soils Area IV and the Northern Buffer Zone. Area Iv Radiological Study Santa Susana Field Laboratory Ventura County, California, Dec. 21, 2012.

Evangeliou, N., et al., 2015. Fire evolution in the radioactive forests of Ukraine and Belarus: future risks for the population and the environment. Ecol. Monogr. 85 (1), 49–72. https://doi.org/10.1890/14-1227.1.

Ewing, 1999. E.C., Nuclear waste forms for actinides. Proc. Natl. Acad. Sci. U.S.A. 96 (7), 3432–3439. https://doi.org/10.1073/pnas.96.7.3432.

Hayaton, 2019. Economic analysis of thorium extraction from monazite. Nuclear Engineering and Technology 51, 631–640, 2019.

IAEA, 2016. International atomic Energy agency/international nuclear information System, citizen-science: potential new opportunity for consent-based siting processes. URL accessed 5/4/2021: https://inis.iaea.org/search/search.aspx?orig_q=RN:50083709.

Japan MOE, 2021. ministry of environment, environment agency, internal and external exposure. URL accessed 8/3/2021. https://www.env.go.jp/en/chemi/rhm/basic-info/1st/pdf/basic-1st-02.pdf.

Kaltofen, 2018. Radioactively-hot particles detected in dusts and soils from Northern Japan by combination of gamma spectrometry, autoradiography, and SEM/EDS analysis and implications in radiation risk assessment. Sci. Total Environ. 1065–1072. https://doi.org/10.1016/j.scitotenv.2017.07.091, 2017 Dec 31;607-608, Epub 2017 Jul 27.

Kaltofen, 2019. Microanalysis of particle-based uranium, thorium, and plutonium in nuclear workers' house dusts. J. Environ. Eng. Sci. https://doi.org/10.1089/ees.2018.0036.

Kaltofen, et al., 2020. Radioactive isotopes measured at olympic and paralympic venues in Fukushima prefecture and tokyo, Japan. J. Environ. Eng. Sci. https://doi.org/10.1089/ees.2020.0139.

LA County, 2019. Los Angeles county after action review of the Woolsey fire incident. URL (accessed 1/4/2021). https://lacounty.gov/wp-content/uploads/Citygate-After-Action-Review-of-the-Woolsey-Fire-Incident-11-17-19.pdf. section 2.1.

LA Times, 2018. Los Angeles times newspaper. URL (accessed 9/18/2020). https://www.latimes.com/projects/la-me-malibu-woolsey-destruction-map/.

Los Alamos, 2000. A Special Edition of the SWEIS Yearbook – Wildfire 2000. Los Alamos National Laboratory. LA-UR-00-3471.

Los Alamos, 2001. Storm Water Quality in Los Alamos Canyon Following the Cerro Grande Fire. Los Alamos National Laboratory. LA-13816-MS.

Mainichi, 2017. Wildfire rages in highly radioactive Fukushima mountain forest. URL accessed 4/30/21. https://mainichi.jp/english/articles/20170501/p2a/00m/0na/003000c.

Nature (2018) 562, 480-482 (doi: https://doi.org/10.1038/d41586-018-07106-5).

NASA, 2018. NASA earth observatory image by joshua stevens, data from NASA/GSFC/METI/ERSDAC/JAROS, national weather service wind speed & direction data, and ASTER science team. URL accessed 8/12/21. https://earthobservatory.nasa.gov/images/144277/woolsey-fire-burn-scar.

Paschoa, A.S., Steinhäusler, F., Paschoa, A.S., Steinhäusler, F., 2010. Chapter 3 - terrestrial, atmospheric, and aquatic natural radioactivity. In: Radioactivity in the Environment, 17. Elsevier, pp. 29–85. https://doi.org/10.1016/S1569-4860(09)01703-3. ISSN 1569-4860, ISBN 9780080449364.

PVT citing, B.L.M., 2021. Pahrump valley times, wildfire at Nevada national security site nears a known contamination zone, 5/20/2021, citing United States bureau of land management and Nevada national security site. URL accessed 5/24/21. https://pvtimes.com/news/wildfire-at-nevada-national-security-site-nears-a-known-contamination-zone-99370/.

Safecast, 2017. Citing mainichi shimbun. URL accessed 4/30/21. https://safecast.org/2017/05/mount-juman-forest-fire-in-namie/.

Silverton, J, et al., 2011. Citizen-science reveals unexpected continental-scale evolutionary change in a Model organism. PLoS One 6 (4), e18927. https://doi.org/10.1371/journal.pone.0018927.

Tew, 1968. Engineering Specifications, Thorium Oxide Target Elements. Douglas United Nuclear, Inc., 4/22/1968, RL-REA-2177.

US DOE, 2005. Historical site assessment of area IV Santa Susana field laboratory Ventura county, California. Methodology, URL (accessed 12/1/20). https://www.etec.energy.gov/Library/Main/SSFLAreaIVHSAVolume%201.pdf, 1.

US DOE (2020a) URL (accessed 11/19/20) https://www.etec.energy.gov/library/Main/sre-C.pdf.

US DOE (2020b) URL (accessed 11/19/20) https://www.etec.energy.gov/Community_Involvement/Newsletters.php.

USC, 2021. United States Code, Title-15 §3724 (B2) and (B3), Crowdsourcing and Citizen-Science.

WADOH, 2008. Washington Department of Health, Environmental Monitoring and Assessment Program, 2007 Hanford/Wautoma Wildfire Summary. DOH/320-048.