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TECHNICAL PAPER

Cesium emissions from laboratory fires

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ABSTRACT

If a radiological incident such as a nuclear power plant accident, a radiological dispersal device, or detonation of an improvised nuclear device occurs, significant areas may be contaminated. Initial cleanup priorities would likely focus on populated areas, leaving the forested areas to pass several seasons where the overhead canopy materials would fall to the forest floor. In the event of a wildfire in a radionuclide-contaminated forest, some radionuclides would be emitted in the air while the rest would remain in the ash. This paper reports on a laboratory simulation study that examines the partitioning of cesium-133 (a nonradioactive isotope of cesium) between airborne particulate matter and residual nonentrained ash when pine needles and peat are doped with cesium. Only 1–2.5% of the doped cesium in pine needles was emitted as particulate matter, and most of the cesium was concentrated in the particulate fraction greater than 10 μm in aerodynamic diameter. For peat fires, virtually all of the cesium remained in the ash. The results from this study will be used for modeling efforts to assess potential exposure risks to firefighters and the surrounding public.

Implications: There is a potential for emissions of radionuclides such as cesium-137 from a wildfire over a radionuclide-contaminated forest. This paper reports on a laboratory simulation study of a wildfire with two types of biomass doped with nonradioactive cesium. This simulation suggests that only 1–2.5% of the cesium in the biomass will be emitted from the wildfire, while the rest will reside in the residual ash. In this study, pine needles were the only contributor to the air emissions of cesium; duff was not a source of cesium emissions. In this study, cesium emitted from the simulated wildfire was concentrated in the particle sizes larger than 10 μm.

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Introduction

Following a radiological release event (i.e., radiological dispersal device [RDD], nuclear power plant [NPP] incident, improvised nuclear device [IND], nuclear testing site, or hazardous waste site) a wide area may be contaminated by radiological materials, including significant forest areas.

The largest historical NPP accidents, the Chernobyl NPP accident near Pripyat, Ukraine (April 1986), and the Fukushima Daiichi NPP accident in the Fukushima Prefecture, Japan (March 2011), released large amounts of various radionuclides into the environment. The International Atomic Energy Agency (IAEA) rated both major accidents the highest level, 7, on the International Nuclear Event Scale (IAEA 2011). Both accidents resulted in major releases of volatile radionuclides, including noble gases, iodine, cesium, strontium, tellurium, and plutonium (Steinhauser, Brandl, and Johnson 2014). The most heavily contaminated areas were evacuated, establishing these areas as "exclusion zones" (EZs). Forest was the main area of contamination for both accidents (Yoschenko et al. 2017). The Chernobyl EZ is estimated to be 4300 km^2 and comprises 70% forest and 30% agricultural land (IAEA 1996; Evangeliou et al. 2015; Hao 2009; Steinhauser, Brandl, and Johnson 2014). The initial Fukushima EZ was much smaller at 600 km^2 (Steinhauser 2014). The contaminated forest was approximately 71% of the impacted Fukushima Prefecture. Approximately 428 km^2 of forest was contaminated with radioactive cesium (^{137}Cs) at a level higher than 1 MBq/m^2 in November 2011 (IAEA 2011).

Initial cleanup priorities after an incident would likely focus on populated areas, leaving the forested areas to pass for one or more seasons, where the overhead canopy materials (where initial deposition of

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radionuclides would occur) would fall to the forest floor. The nuclear power plant (NPP) accidents in Chernobyl and Fukushima showed that the contaminated forests were not remediated on a large scale due to lack of fast and cost-effective mitigation methods (Hashimoto et al. 2012; Jouve et al. 1993; Linkov, Morel, and Schell 1997). Forest decontamination is difficult because modification of forest via decontamination (e.g., topsoil removal) may disturb forest ecosystems via erosion, sedimentation, disruption of habitat, and loss of nutrients (Jouve et al. 1993).

Cesium-137 is the most significant radionuclide contaminant released during radiological incidents. Cesium-137 is a high-yield contaminant with a long half-life of 30.2 years, is water soluble, and is easily taken up by plants as a biological analog to potassium (Garten et al. 2000; Paliouris et al. 1994; Saffarzadeh et al. 2014; Yoschenko et al. 2017). Long after the primary atmospheric ^{137}Cs has dissipated from contaminated areas, high levels of ^{137}Cs have been found in vegetation, litter, and concentrated in the top 5 cm of soil (Hejl et al. 2012; Paliouris et al. 1994; Yoschenko et al. 2006). This secondary contamination has been found to be redistributed into the atmosphere by forest fires (Bourcier et al. 2010; Commodore et al. 2012; Dusha-Gudym 2005; Evangeliou et al. 2014; Hao 2009; Paatero et al. 2009; Paliouris et al. 1994; Strode et al. 2012; Wotawa et al. 2006; Yoschenko et al. 2006). Based on cesium material balance studies on ash, during a typical biomass fire, 40–70% of cesium in the fuels can be released in the atmosphere with increased release as fire temperatures rise (Amiro et al. 1996). For these reasons, Cs was chosen to be the contaminant of choice for this study.

Wildfires have the potential to be a significant source of secondary contamination and exposure during and following the initial response. A large wildfire may generate large amounts of radioactive materials suspended in the atmosphere to cause exposure risk to firefighters and the nearby public and contaminate or recontaminate wide areas by atmospheric dispersion (Kashparov et al. 2000; Hohl et al. 2012; Evangeliou et al. 2014; Viner et al. 2015; Goldammer, Statheropoulos, and Andreae 2009; Goldammer et al. 2017). Inhalation of even small amounts of radioactive material has the potential to raise the risk of developing cancer later in life. Research is needed (1) to assess the exposure risk of firefighters and the public from a wildfire; (2) to predict secondary contamination potential due to wildfires in the radiologically contaminated forest; and (3) to develop appropriate forest management strategies for these radionuclide-contaminated forest areas. Research findings can provide critical information for responders and nearby communities to help prepare for a large fire in the radioactively contaminated forest and can aid in development of effective management strategies for contaminated forests.

To address this potential environmental health threat, a collaborative research effort was carried out between the U. S. Environmental Protection Agency and the U.S. Forest Service to simulate emissions in laboratory fires of pine needles and duff doped with nonradioactive cesium. The objective was to determine the partitioning of Cs between airborne particulate and residual ash from burning Cs-contaminated vegetation as a function of particle size distribution (PSD), and to correlate Cs emissions with carbon-containing compounds and combustion efficiency. The type of vegetation burned was similar to fire-prone areas where nuclear facilities exist in the western United States. Two examples would be Los Alamos, NM, and Hanford, WA. Cesium (Cs-133), a nonradioactive Cs isotope, was selected as a surrogate radioactive material for this study (Amiro et al. 1996). The nonradioactive isotope of Cs will chemically behave identically to the radioactive isotopes, although quantification of Cs in the biomass and stack samples will need to use nonradioactive detection methods such as x-ray fluorescence (XRF) or inductively coupled plasma–mass spectrometry (ICP-MS). The study results then can be used to assess the exposure risk and potential secondary environmental contamination due to wildfire in a radionuclide-contaminated forest.

Materials and methods

Material

Two different biomass materials were selected based on likely western U.S. forest fire settings where NPP accidents and wildfires might occur. One biomass fuel represented the materials on the forest floor (pine needles) and the other biomass fuel represented the forest floor itself (peat moss simulating the duff layer), to simulate flaming as well as smoldering combustion of a wildfire.

The biomass was doped with nonradioactive cesium chloride (CsCl) to test the effects on emissions of biomass contamination from a nuclear power plant accident. CsCl was selected as the chemical form of Cs for these tests for several reasons: (1) CsCl is a common form of Cs for large radioactive sources (either Cs-134 or Cs-137); (2) the Department of Homeland Security's National Planning Scenario 11 and various national-level exercises examining response and recovery from radiological dispersal device incidents used CsCl as the chemical form of the contaminant (EPA 2010; 2012;

FEMA 2017; Steuteville 2010); (3) CsCl sources have been involved in incidents involving fatalities due to radiation exposure (IAEA 1988); and (4) the water-soluble nature of CsCl facilitates distribution of the contaminant in experimental materials.

Target initial biomass concentrations of the added CsCl (Sigma Aldrich, St. Louis, MO) were calculated based on method detection limits of the analytical instruments to be used (XRF, ICP-MS). Cesium concentrations for this simulation were likely several orders of magnitude higher than would likely be found from an actual radiological incident, but since gamma spectroscopy is not an option for non-radioactive Cs, the initial target concentrations for the biomass materials needed to be high enough so that the experimental results could be detected. It is not expected that this experimental consideration would result in any statistically significant uncertainties because despite the difference between the amounts used for the experiment and the amount anticipated from a real incident, the Cs would be expected to distribute through the same mechanisms. Even with the artificially increased Cs concentrations, Cs was still present in much lower concentrations than other alkali metals such as sodium and potassium.

Each fuel was doped with an aqueous solution of CsCl to achieve a concentration of 1.5 g Cs/kg vegetation. Batches of 2 kg pine needles were misted with 400 mL of a 9.5-g/L CsCl aqueous solution from a nebulizer while being rotated and mixed in a 246-L compost tumbler. A total of 16 kg pine needles was doped with CsCl for triplicate experimental burns. After the CsCl solution was applied, the needles were dried in a conditioning chamber for 48 hr at 30°C and 20% relative humidity (RH) in large aluminum pans. Figure 1a shows a photograph of the prepared pine needles.

The peat was doped by spraying a 2-kg batch with 600 mL of the CsCl solution in an aluminum pan, mixed well, and then transferred to the composter for tumbling and mixing. The doped peat was subsequently transferred to aluminum pans and conditioned in a drying oven at 85°C for 24 hr. A total of 8 kg peat was doped with CsCl. Figure 1b shows a photograph of the prepared peat.

Fire test setup

The combustion chamber where the experiment was conducted at the Missoula Fire Sciences Laboratory is a square room measuring 12.4 m × 12.4 m × 19.6 m high with a total volume of ~3000 m^3 (see Figure 2). An exhaust stack located at the center of the room extends from 2.1 m above the floor and extends up to the ceiling of the chamber. An inverted funnel at the bottom of the exhaust stack narrows from a 3.6-m diameter opening to the 1.6-m stack diameter sampling ports that pass through the walls of the exhaust stack, located ~15.5 m above the floor, and accessed from a platform built around the stack near the ceiling of the chamber.

The burn chamber temperature was set at 26.7°C and the RH at 25%. The mean flow velocities in the exhaust stack were 3.7–4.6 m/sec, to provide an adequate volume for the filter samplers.

Each fuel was burned in triplicate, for a total of six tests with doped biomass, plus two blank runs, one with each of the undoped biomass fuels.

The fuel bed for the pine needles was on a ceramic board 61 cm wide × 122 cm long, inside a frame of concrete blocks, and was positioned on the floor centered below the stack. Initially, one bag of approximately 500 g pine needles was manually spread out on the bed and ignited with a lighter at one end with a small amount of ethanol from a squirt bottle. As the flame front burned the length of the fuel bed to the other end, an additional 500 g pine needles was manually added from another bag, and the fire propagated back to the initial end. This procedure was repeated for 10 to 12 bags for a total of ~5 kg fuel. Each bag of pine needles was weighed immediately before addition to the

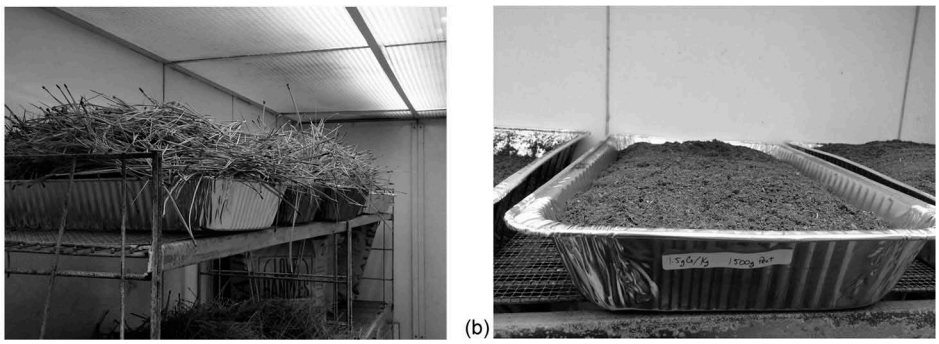

Figure 1. (a) Prepared pine needles; (b) prepared peat.

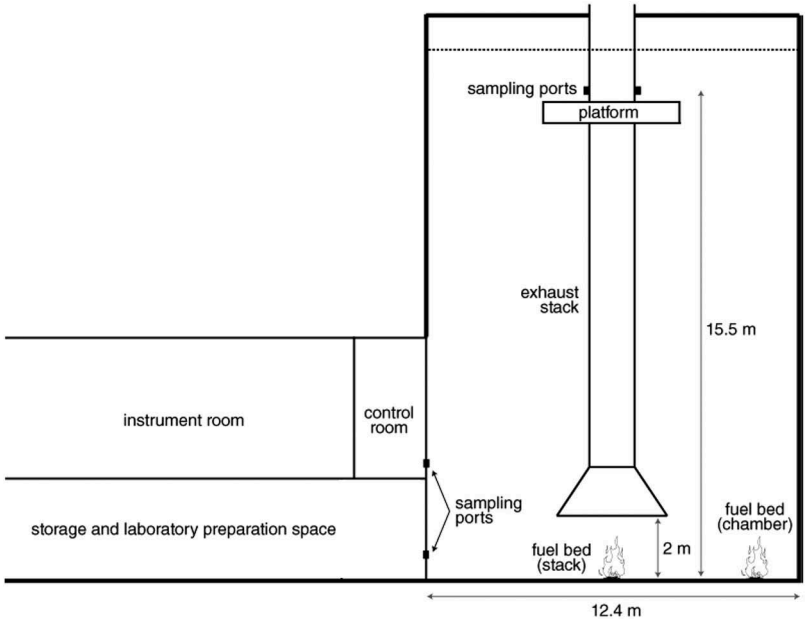

Figure 2. Missoula Fire Sciences laboratory burn chamber.

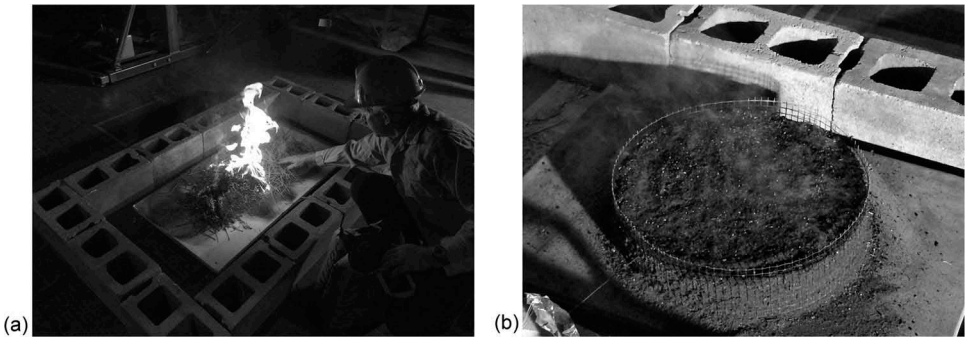

Figure 3. (a) Pine-needle burn; (b) peat burn.

fuel bed. Figure 3a is a photograph of a pine-needle combustion test and, each experiment lasted approximately 1 hr (Table 1).

For the peat moss fuel beds, 2–3 kg of peat was packed into a 50 cm diameter × 12 cm height circular form made from metal hardware screen (1 cm mesh) to construct a simulated duff layer sample. The peat moss fuel bed was placed on the ceramic board and ignited with a propane torch until glowing combustion was initiated around the surface and visible smoke was emitted. A fuel moisture sample was collected from the fuel bed in a metal can prior to each of the needle test fires. Additional moisture samples were collected at subsequent 15-minintervals from the supply of needles that were about to be added to the fire. A single moisture sample of the peat was collected as the circular peat bed was assembled just prior to ignition. Figure 3b is a photograph of a peat burn, which lasted for approximately 2 hr (Table 1).

Each run had a data sheet with sample container identifications (IDs), ignition and end times, fuel weights, and fire observations. A picture of each fuel bed was made before ignition with a sign for run ID. The run end time was determined based on pump flow rates and sample volumes specified by the respective particulate matter (PM) filter sampling methods. The pine-needle ash was allowed to cool, collected in aluminum pans, and weighed. The residual unburned peat and ash was covered with foil to halt combustion and cool before transfer to pans and weighing.

Ash subsamples were later shipped to the EPA's facilities in Research Triangle Park, NC, for chemical analysis by XRF and ICP-MS. Sample chain of custody forms were used to assure that the samples and their respective tests remained linked.

In total, eight experimental burns took place. The blank run of each fuel not doped with Cs was conducted prior to the burns involving biomass doped with Cs.

Table 1. Fuels, chamber conditions, and burn data.

Run	Date	Fuel	Type	Run duration (hr:min)	Chamber temperature (°C)	Chamber RH (%)	Fuel moisture (%)	Fuel weight (kg)	Ash weight (kg)	Fuel consumption (%)
1	3/22/16	Pine needles	Blank	1:00	26.1	19.5	8.4	5.00	0.20	96.0
2	3/22/16	Peat	Blank	1:34	26.4	15.6	24.9	2.92	1.42	51.5
3	3/23/16	Peat	Cs doped	2:01	26.0	19.1	30.0	2.70	1.36	49.6
4	3/23/16	Peat	Cs doped	2:00	28.1	16.4	78.2	2.40	1.11	53.5
5	3/23/16	Peat	Cs doped	2:00	27.4	13.9	34.1	2.90	1.53	47.1
6	3/24/16	Pine needles	Cs doped	1:01	24.4	23.3	5.6	5.55	0.36	93.5
7	3/24/16	Pine needles	Cs doped	1:01	24.9	22.4	5.4	5.24	0.31	94.0
8	3/24/16	Pine needles	Cs doped	1:01	24.3	19.5	6.5	5.16	0.38	92.6

Continuous emission monitors (CEMs)

Carbon gases (CO_2, CO, and CH_4) were continuously measured from the stack platform with a Picarro G2401-m CRDS gas analyzer (Picarro, Santa Clara, CA). The analyzer has a response time of <2.5 sec, and an external pump with a flow rate of 0.6 L/min. The instrument was calibrated before and after each run with three calibration gas mixes with a range of CO (0.092, 3.03, 9.75 ppm), CH_4 (1.49, 3.04, 4.96 ppm), and CO_2 (351, 510, 745 ppm) concentration in each mix. A stainless-steel sample probe for the CEM was inserted approximately 1 m into the stack. The real-time monitoring of these gases provides an assessment of the reproducibility of the fire conditions and their potential impact on the particulate samples taken in the stack, given that the particulate samples are integrated over an extended period of time and do not provide an indication of the instantaneous behavior of the fire during the sampling periods. The measurement of the primary carbon emissions (CO_2, CO, and CH_4) provides increased assurance that the particulate measurements were taken under similar combustion conditions for each fire.

Canisters

Canister samples were collected from the stack using the CEM sample line at 15- to 20-min intervals during the burns with a 4-L/min KNF pump (KNF Neuberger, Trenton, NJ). SUMMA canisters (850 mL) were filled to approximately 172.4 kPa (25 psia). The concentrations of CO, CO_2, CH_4, and light hydrocarbons in the canisters were measured with an Agilent model 7890 gas chromatograph (Agilent Technologies, Santa Clara, CA) equipped with dual flame ionization detectors (300°C). Analysis for CO and CO_2 utilized a 0.32 cm diameter × 183 cm long Carbosphere column (Grace Alltech, Columbia, MD) with helium carrier gas at a flow rate of 16 mL/min. The oven temperature program was 50°C for 6 min, 10°C/min to 130°C, 130°C for 6 min, with a nickel catalyst methanizer (375 °C) to convert the CO and CO_2 to methane for detection by the flame ionization detector (FID). The analysis of CH_4 occurred simultaneously on the other FID with a 50-m HP/ALS 0.53 mm mega-bore capillary column (Agilent Technologies, Santa Clara, CA), with helium carrier gas at 6 mL/min. A set of gas standards bracketing the sample concentrations was analyzed to calibrate the instrument for each compound. Duplicate sample analyses were performed every sixth sample. National Institute for Standards and Technology primary standards for CO, CO_2, and CH_4 were analyzed daily to validate the standard curves.

Particulate samples

U.S, Environmental Protection Agency reference Methods 5 (EPA 1996a) and 201A (EPA 1996b) sampling was conducted simultaneously through port openings on the stack platform located 15.5 m above the fuel bed. Method 5 was used to determine the total filterable particulate matter (PM) from the source. The exhaust stream was sampled along a cross section of the stack and PM was captured within the nozzle, probe, filter bell, and on the 0.3-μm pore size glass-fiber filter. The filter was maintained at a temperature of 120 ± 4°C followed by impingers maintained at a temperature below 18°C. For Method 201A, PM_{10} and $PM_{2.5}$ cyclones were run in tandem to achieve multiple aerosol cut points.

Fuel, filter, and ash analysis

The Cs content from samples (biomass fuels, ashes, PM filters, and probe rinse) was analyzed using XRF and ICP-MS. XRF analyses were performed in-house at the EPA facilities in Research Triangle Park, NC. ICP-MS analyses were performed by a commercial laboratory (TestAmerica, Earth City, NJ).

The pine-needle samples were first processed with an electric grinder to create a more homogeneous sample. A 2-g portion was taken and further ground using a mortar and pestle while adding 2 mL liquid binder,

0.1 g $[-C_{10}H_9ON-]_n$ per 1 mL methylene chloride, grinding until all of the liquid binder had evaporated. The powder was then added to a die and placed into a press under vacuum, where it was forced into a useable pellet. The peat samples were found to be sufficiently fine that they did not need to be ground in the electric grinder. The filter samples all had a 42-mm center section punched out for fitting in the sample holder cups. All of the samples were secured on a polypropylene film inside a sample cup for analysis.

The XRF analysis was performed on a Philips PW2404 XRF instrument (Philips, Almelo, The Netherlands) using the IQ+ Cs method contained in the instrument's software. The IQ+ Cs method specifically determines all of the elements over a full 10 scans and then provides 10 sec of counting time for two cesium analytical lines to increase sensitivity. The resulting scans were then reviewed with interference lines removed. The pellet samples and filter samples were calculated as their constituent elements because the software was unable to differentiate between compounds containing the same element. The filter sample results were normalized to 100% assuming all of the detected fluorine to be part of the Teflon filter; the pellet results were not normalized to take into account the large amount of carbon that is invisible to the detector.

For ICP-MS analysis, all samples except probe rinse samples were digested using EPA Method 3050B (EPA 1996c). The extracted samples were then analyzed for Cs using the ICP-MS following EPA Method 6020a (EPA 1998). Each sample was analyzed to achieve the lowest possible detection limit within the constraints of the method. In some cases, due to interference or analytes present at high concentrations, samples were diluted. For diluted samples, the detection limits were adjusted relative to the dilution required. Calculations were performed before rounding to avoid round-off errors in calculated results. All holding times were met, and proper preservation noted for the methods was performed on these samples.

Results

Burn results

Durations, chamber conditions, and fuel parameters are listed in Table 1. The pine-needle burns were 1 hr long, which was sufficient to consume 5 kg needles and collect adequate stack gas volumes for the filter samples on the Method 201A and Method 5 trains. Approximately 95% of the fuel mass was consumed in each of the pine-needle burns. The Cs-doped pine needles had very low fuel moisture content (5–6%), which would be extreme conditions for a wildfire. Two of the peat fuel beds were dry (25–30%), and Run 4 peat was much moister, 78.2%. This difference in moisture content did not seem to affect the total consumption, as the three peat burns consumed 47.1 to 53.5% of the biomass over the 2-hr burn duration.

CEM results

Results for the Picarro CEM are given in Figure 4. Levels of CO, CH_4, and CO_2 were low, just slightly above background concentrations. Run 1, the pine-needle blank, had a brief increase in CO_2, indicating flaming combustion. The high flow rate up the stack that was needed to provide an adequate volume for the filter samplers may be the reason that trace gas concentrations were very low. Other burns that had peaks of CO_2 detected by the Picarro CEM were Runs 2, 4, and 5. The concentrations of CO and CH_4 for all of the runs were low, without measurable trace levels detected above background for many of the runs.

Canister results

The canister results (Table 2), like the CEM data, contained very low concentrations of the gases measured. Light hydrocarbons (C_2–C_5 compounds) were below the limits of detection and are not reported. The results for all of the runs are similar in concentration range. Levels of CO, an indicator of smoldering, ranged from 0.16 to 0.22 ppm for the pine needles and from 0.17 to 0.44 ppm for the peat.

Emission factors

Fire-integrated emission factors were calculated using the carbon mass balance method (Ward and Radke 1993). The emission factors are reported in Table 3. The emission factors for all eight runs for CO, CH_4, and CO_2 were indicative of high-efficiency combustion, as were the modified combustion efficiency (MCE) values (Ward and Radke 1993), which were in a range of 0.97–0.99 due to the low trace levels of gases measured in the diluted stack. The CO_2 emission factors were higher for the pine-needle runs than for the peat runs, in a range of 1795–1809 g/kg and 1746–1806 g/kg, respectively. CO emission factors were higher for the peat than for the pine needles, indicative of greater smoldering combustion. The CH_4 emission factor (11.5 g/kg) for peat Run 5 was more than twice that

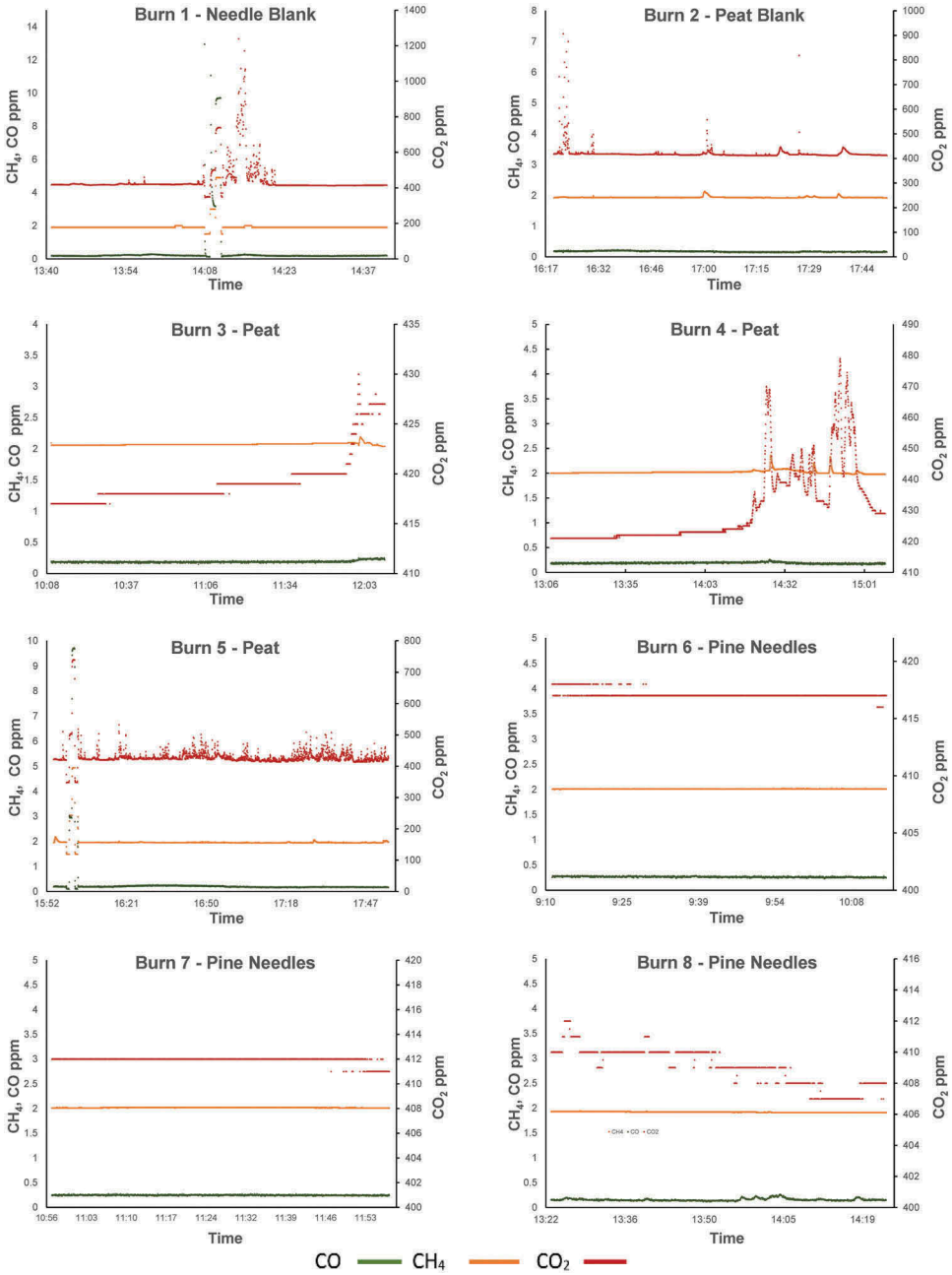

Figure 4. Stack CO, CH_4, and CO_2 concentrations (ppm) from Picarro G2401-m CEM.

of any other run. Run 5 (peat) also had the highest CO emission factor (35.5 g/kg).

Fuel and ash analysis

Table 4 lists the results from the analysis of the biomass and the ash for Cs. In general, approximately 40–50% of the Cs doped into the biomass was recovered. There are several possible reasons for this discrepancy, including (1) uncertainty in the XRF analyses, considering that XRF results assume that the Cs in the ash is in the form of Cs_2O, which may or may not be true; and (2) assumptions that the Cs was uniformly distributed in the ash sample, which is unknown, although efforts were made to ensure that Cs was evenly distributed in the original biomass (using the spray bottle and rotating composting bin to mix the dopant and the biomass).

Results from extractive stack samples

Table 5 lists the PM results from the stack sampling Method 5 and Method 201A trains. The O-ring on one

Table 2. Concentrations of CO, CH_4, and CO_2 (ppm) in canister samples.

Run	Date	Time (MST)	Fuel	CO	CO_2	CH_4
1	3/22/16	13:52	Pine-needle blank	0.34	415	2.10
1	3/22/16	14:01	Pine-needle blank	0.34	414	2.10
1	3/22/16	14:17	Pine-needle blank	0.23	414	2.05
1	3/22/16	14:36	Pine-needle blank	0.23	410	2.10
2	3/22/16	16:00	Background	0.17	401	2.05
2	3/22/16	16:23	Peat blank	0.16	410	2.12
2	3/22/16	16:55	Peat blank	0.16	415	2.07
2	3/22/16	17:13	Peat blank	0.21	404	2.07
2	3/22/16	17:45	Peat blank	0.16	403	2.07
3	3/23/16	10:10	Cs-doped peat	0.23	420	2.07
3	3/23/16	10:51	Cs-doped peat	0.38	418	2.07
3	3/23/16	11:19	Cs-doped peat	0.18	426	2.12
3	3/23/16	11:45	Cs-doped peat	0.17	415	2.12
3	3/23/16	12:05	Cs-doped peat	0.20	414	2.12
4	3/23/16	13:10	Cs-doped peat	0.21	409	2.07
4	3/23/16	14:10	Cs-doped peat	0.17	412	2.07
4	3/23/16	14:40	Cs-doped peat	0.33	411	2.07
5	3/23/16	16:00	Cs-doped peat	0.44	409	2.05
5	3/23/16	16:30	Cs-doped peat	0.19	407	2.26
5	3/23/16	17:51	Cs-doped peat	0.17	407	2.05
6	3/24/16	9:00	Background	0.15	405	2.05
6	3/24/16	9:17	Cs-doped pine needles	0.18	408	2.05
6	3/24/16	9:35	Cs-doped pine needles	0.20	407	2.08
6	3/24/16	9:55	Cs-doped pine needles	0.20	410	2.14
6	3/24/16	10:10	Cs-doped pine needles	0.18	424	2.14
7	3/24/16	11:03	Cs-doped pine needles	0.18	421	2.08
7	3/24/16	11:18	Cs-doped pine needles	0.17	408	2.08
7	3/24/16	11:36	Cs-doped pine needles	0.21	410	2.08
7	3/24/16	11:55	Cs-doped pine needles	0.17	412	2.05
8	3/24/16	13:26	Cs-doped pine needles	0.16	409	2.05
8	3/24/16	13:42	Cs-doped pine needles	0.20	407	2.11
8	3/24/16	14:05	Cs-doped pine needles	0.22	415	2.11

Note. MST: Mountain Standard Time.

Table 3. Integrated emission factors (g/kg) and modified combustion efficiency (MCE) from canister results.

Run	Fuel	Data	CO	CO_2	CH_4	MCE
1	Pine-needle blank	Mean	22.8	1789	3.1	0.98
		s.d.	5.6	14.5	2.4	0.00
2	Peat blank	Mean	22.4	1785	4.8	0.98
		s.d.	19.7	35.3	2.5	0.02
3	Cs-doped peat	Mean	20.4	1786	5.5	0.98
		s.d.	13.0	23.9	4.7	0.01
4	Cs-doped peat	Mean	14.8	1806	1.4	0.99
		s.d.	8.0	12.6	0.3	0.01
5	Cs-doped peat	Mean	35.5	1746	11.5	0.97
		s.d.	24.5	51.7	19.4	0.02
6	Cs-doped pine needles	Mean	17.0	1797	3.4	0.99
		s.d.	7.9	16.5	3.4	0.01
7	Cs-doped pine needles	Mean	11.6	1809	2.2	0.99
		s.d.	5.9	13.9	1.8	0.01
8	Cs-doped pine needles	Mean	17.1	1795	4.1	0.99
		s.d.	10.3	28.2	4.5	0.01

Note. s.d., standard deviation.

of the Method 201A cyclones failed on Run 1, so data for this fraction for Run 1 were not usable.

Table 6 lists the results of cumulative Cs mass air emission from the Method 5 train, based on using XRF to analyze the filters from the train and using ICP-MS to analyze the probe rinse results. The Cs results from the Method 5 filters were very close to the method detection limit and under the quantitation limit for the XRF. These results were what suggested using the more sensitive ICP-MS method for subsequent analyses. No Cs was observed in the probe rinse for any of the peat burn samples.

Table 4. Fuel and ash analysis (via XRF).

Run	Initial fuel mass (kg)	Initial Cs (% mass)	Initial Cs (g)	Final ash mass (kg)	Cs in ash (% mass as Cs_2O)	Cs in ash (g)
1	5.0	0.01	0.7	0.20	0.04	0.07
2	2.9	0.01	0.4	1.42	0.004	0.05
3	2.7	0.52	14.0	1.36	0.41	5.29
4	2.4	0.43	10.4	1.12	0.52	5.46
5	2.9	0.48	13.9	1.53	0.38	5.50
6	5.6	0.17	9.6	0.36	1.47	4.98
7	5.2	0.25	13.3	0.32	1.48	4.39
8	5.2	0.21	10.8	0.38	1.31	4.73

Table 5. Stack sampling data.

Run	Stack flow (dscm/min)	Run time (min)	Total stack volume (dscm)	M5 PM (g)	M5 PM (mg/dscm)	M201A Total PM (g)	M201A PM (mg/dscm)
1	389	60	23365	2.6E-03	2.46	9.3E-03	N/A
2	350	90	31459	8.0E-04	0.56	3.3E-03	4.16
3	395	120	47358	1.3E-03	0.60	6.2E-03	4.15
4	364	120	43681	4.0E-04	0.20	4.8E-03	3.21
5	386	120	46322	1.1E-03	0.53	4.4E-03	2.95
6	288	60	17252	1.4E-03	1.76	6.0E-03	8.81
7	388	60	23271	2.4E-03	2.24	4.2E-03	5.61
8	450	60	26980	2.0E-03	1.62	4.5E-03	6.04

Note. N/A, not available—O-ring failed on cyclone.

Table 6. Cumulative Cs mass air emission results.

Run	Cs (g) (filters)	Cs (g) (probe rinse)	Total Cs mass (g)
1	ND	ND	ND
2	2.16E-03	ND	2.16E-03
3	3.48E-03	ND	3.48E-03
4	9.89E-04	ND	9.89E-04
5	ND	ND	ND
6	1.43E-03	7.89E-03	9.33E-03
7	7.38E-03	3.18E-02	3.92E-02
8	5.77E-03	4.81E-03	1.06E-02

Note. ND, not detected.

Table 7 lists the summary results for the duff burns, while Table 8 lists the summary results for the pine-needle burns. The issue for the high reported Cs mass in the peat blank run 2 sample as well as several other of the blank samples comes from several factors. Between the increased sensitivity of the detector at the two Cs analytical lines and the normalization to take the filter mass into account, the 0.012% Cs should be interpreted as biased noise, reinforced by the 0-g reported sample mass being under the desired limit for the XRF. In addition, the majority of blanks that are reported to have Cs contain roughly the same percentage of Cs, suggesting that there is no Cs at detectable levels in these samples. The XRF results from run 2 (duff blank) suggested that approximately 4% of the Cs was emitted up the stack with the particulate matter, but given that there was no Cs added to the duff material for that test (the tests including doped duff all showed $\ll$1% of the Cs was emitted up the stack) and that the ICP-MS did not show any Cs in the

Table 7. Summary of duff results.

				XRF analysis			ICP-MS analysis		
Run	Fuel burned (kg)	Cs in ash (g)	Cs in ash (g/kg burned)	Cs in PM (g)	Cs in PM (g/kg burned)	Percent Cs emitted	Cs in PM (g)	Cs in PM (g/kg burned)	Percent Cs emitted
2	1.5	0.1	0.04	2.16E-03	1.44E-03	3.89	ND	ND	ND
3	1.3	5.3	3.94	3.48E-03	2.60E-03	0.07	ND	ND	ND
4	1.3	5.5	4.25	9.89E-04	7.70E-04	0.02	ND	ND	ND
5	1.4	5.5	4.03	ND	ND	ND	ND	ND	ND

Note. ND, not detected.

Table 8. Summary of pine-needle results.

				XRF analysis			ICP-MS analysis		
Run	Fuel burned (kg)	Cs in ash (g)	Cs in ash (g/kg burned)	Cs in PM (g)	Cs in PM (g/kg burned)	Percent Cs emitted	Cs in PM (g)	Cs in PM (g/kg burned)	Percent Cs emitted
1	4.8	0.1	0.01	ND	ND	ND	ND	ND	ND
6	5.2	5.0	0.96	1.43E-03	2.76E-04	0.03	5.84E-02	1.12E-02	1.16
7	4.9	4.4	0.89	7.38E-03	1.50E-03	0.17	1.19E-01	2.41E-02	2.64
8	4.8	4.7	0.99	5.77E-03	1.21E-03	0.12	6.03E-02	1.26E-02	1.26

Note. ND, not detected.

stack emissions for run 2, we believe that the run 2 duff blank sample was reported high due to the very low sample mass, the small quantity of Cs present, and the algorithms that the instrument uses to estimate resulting Cs mass in the sample. If the questionable results on the run 2 duff blank are excluded, it appears that, based on the XRF results, ≪0.07% of the Cs from the duff burns becomes entrained into the combustion emissions and emitted up the stack. The pine-needle results based on the XRF suggest that between 0.03 and 0.17% of the Cs is emitted up the stack, whereas the ICP-MS results suggest that between 1.2 and 2.6% of the Cs is emitted up the stack for the pine-needle runs. Nonhomogeneous samples like filters contribute a large amount of error to the analysis, with results varying based on the orientation of the filter in the XRF, sample distribution on the filter, and especially concentration within the PM. For filter samples and other low-mass samples, ICP-MS would be the superior method, because the level of confidence in the ICP-MS results is much higher. Both methods were planned to be used for the experiments based on initial assumptions of Cs partitioning. It turned out that much less of the Cs partitioned into the airborne particulate matter than initial assumptions estimated. Results from both methods are included for completeness; however, the ICP-MS results were primarily used to formulate the conclusions due to the higher level of confidence in the ICP-MS data.

One of the advantages of Method 201A is that there are multiple splits of the sample based on particle size. Results were obtained for particles <2.5 μm in aerodynamic diameter, between 2.5 and 10 μm in aerodynamic diameter, and >10 μm in aerodynamic diameter. Table 9 (based on the ICP-MS analyses) lists the results from these different splits. For one pine-needle run (run 6), 12% of the Cs was in the <2.5 μm split, with the remaining 88% being found in the >10 μm split. The other two pine-needle runs (runs 7 and 8) showed 100% of the Cs in the >10 μm fraction. This observation suggests that wildfires from radionuclide-contaminated forests would not produce radioactive PM of the type that would be transported long distances.

Table 9. Summary of Cs distribution in different particle sizes.

Run	Fuel type	PM2.5 (% mass)	PM10 (% mass)	> PM10 (% mass)
1	Pine-needle blank	ND	ND	ND
2	Peat blank	ND	ND	ND
3	Cs-doped peat	ND	ND	ND
4	Cs-doped peat	ND	ND	ND
5	Cs-doped peat	ND	ND	ND
6	Cs-doped pine needles	0.0044	ND	0.034
7	Cs-doped pine needles	ND	ND	0.091
8	Cs-doped pine needles	ND	ND	0.037

Note. ND, not detected.

Discussion and operational implications

The study results confirmed the potential inhalation exposure risk to firefighters during the response to the wildfire via radioactive PM and impact to the nearby public by the air emission of radioactive PM from a wildfire. In addition, Cs from the highly concentrated radioactive residual ash may be leached and transported to streams following the initial response to firefighting or precipitation events. Radioactively contaminated forest needs special management plans to prepare for a wildfire and also for postwildfire ash maintenance.

First responder: Exposure potential from fly ash and bottom ash

Based on these test results, several suggestions can be made to improve and/or safeguard emergency response actions and planning, for fighting fires involving a contaminated forest/area.

First, the expected concentration of cesium contamination present in a forest (from a previous atmospheric release/event) is low. The total source term (total amount of cesium present) might be very high when counted over the entire affected area (both Fukushima and Chernobyl events created very large exclusion zones with significant amounts of radioactive contamination and involved a large portion [~70%] of forest coverage). Fortunately, the burning of a contaminated forest serves to further lower the concentration initially present at almost any single point, since the source term is slowly diminishing (due to radioactive decay and weathering processes) while the turbulence from the fire serves to spread the contamination over a wider area. This tends to reduce the hazard from radiological exposure, essentially diluting the concentration and reducing the exposure level to any single individual present in the contaminated area. Thus, external exposure rates (the gamma rays penetrate the body from the exterior and may be absorbed in the body, not including ingestion or inhalation) to responders should be no higher than the rates formerly present in the contaminated forest. If total doses to firefighters are of concern, dose estimates including amounts from ingestion or inhalation can be estimated based on these results. Although the air samples were taken at 15.5 m height, which is much higher than the breathing zone of a firefighter, they will still be representative of the airborne distribution of Cs, and large particles that may not have been entrained into the air would be expected to fall back to the ground in the vicinity of the fire. Individual firefighter doses would be significantly mitigated by use of proper turnout gear and personal protective equipment, considering the observed ratio of coarse to fine particulates in this study. If previous survey results (dose rates or contamination levels) are available for the area of concern, this information could guide response activities such as stay times, expected accumulated dose per day or shift, and other parameters such as initial volumes of air to collect for air concentration measurements.

Depending on the amount of information available about the existing contamination levels before a fire, it may be possible to forecast approximate levels of secondary aerosolizing and deposition (cesium from a previous event that has been re-aerosolized and redeposited due to the most recent fire). Based on the results listed in the preceding, for similar fire conditions, it is expected that no more than 1–3% of the existing contamination on the readily combustible material (i.e., the pine needles in this study, which were considered very dry for natural conditions) will be released as airborne emissions. This seeming contradiction of prior published research (Amiro et al. 1996) can be explained by differences in experimental methodology. The previous work was based on Cs mass balances in the ash, whereas the study described in this paper was based on PM emissions taken at a distance (~15.5 m) above the fire. It is entirely possible that more Cs was released from our fires but, due to particle size, was not sufficiently entrained in the upward flow to be measured in the stack. The contamination in the duff layer can be ignored (at least in fires similar to the test conditions—conflagrations with higher temperatures might be expected to liberate some of the contamination in the duff layer). This 1–3% can be used as a possible source term for the secondary release, which (combined with an estimated volume of fire exhaust) might give usable atmospheric concentration estimates. Of course, direct measurements of the actual existing concentration should be performed as soon as possible to reduce the inherent inaccuracies of estimates.

The risk to responders of inhaling airborne contamination should be prevented if the responders wear the usual firefighter turnout gear, which incorporates self-contained breathing apparatus (SCBA). If woodland firefighters do not typically wear SCBA, use of SCBA might be justified in the case of fires involving contaminated forests. An analysis of possible concentrations would inform this case; if the expected concentrations are greater than 10% of the applicable derived air concentration (DAC) for cesium, then breathing zone protection is justified. A DAC is defined by the Nuclear Regulatory Commission as "the concentration of a given radionuclide in air which, if breathed by the reference man for a working year of 2,000 hours under conditions of light work (with an inhalation rate of 1.2 cubic meters of air per hour), results in an intake of one annual limit on intake (ALI) for that nuclide or combination of nuclides." Doses derived from the DAC can be adjusted for moderate or heavy work conditions since these definitions incorporate higher volumes of air breathed per hour. The presence of other radionuclides that might become re-aerosolized must also be considered and guarded against in the same manner. DAC values for various isotopes are available in many health physics references that address inhalation concerns (Federal Register 1994; Schleien 1992). A health

physicist should be consulted when calculating air concentration levels to ensure appropriate calculations and assumptions are used.

The results from this study can be used to identify remediation and forest management options. There are a number of remediation methods that can be applied to manage contaminated forests to lessen the probability or effects of burning. Areas can be severely contaminated by cesium-137, strontium-90, multiple isotopes of plutonium, and more than a dozen other radionuclides (Bird and Little 2013). A basic strategy would be construction of firebreaks and roads as an aid to fire suppression to reduce the area burned if a wildfire occurs. Thinning or cutting of trees will lessen the amount of contaminated biomass that could be consumed by a wildfire. At Chernobyl, a 4-km^2 stand of Scots pines (*Pinus sylvestris*) was felled, bulldozed, and buried—all the trees that were subject to the most severe fallout. A tree-felling device can greatly reduce the human exposure compared to individuals with chain saws. To contain the radionuclides that fell on the zone's waterways, a series of dikes was constructed, designed to prevent flooding into the Pripyat River (Jouve et al. 1996). Trees and other plants fix radionuclides through their basic growth and maintenance processes. When leaves and needles transpire, the plant draws more water up from the roots. The water-soluble salts of cesium are chemical analogs of potassium and calcium, respectively, and are taken up in place of these crucial nutrients. Concentrations of radionuclides gradually accumulate in needles as each season progresses. The needles then fall to the ground, becoming part of the litter, and returning the radioactive salts to the top layer of the soil in a natural cycle that takes approximately 10 to 12 yr to complete (Bird and Little 2013). Contaminated litter can be removed from high impact or other key sites. During a field experiment carried out in 1994 in Belarus, a significant part of the ^{137}Cs associated with the fine fraction of organic matter was still incorporated into the mineral fraction of the soil. It is therefore likely necessary to remove a part of the mineral soil to achieve a significant decontamination factor (Jouve et al. 1996).

Depending on the condition of the affected site, burned areas require urgent and long-term restoration strategies to prevent secondary damage such as landslides and sediment runoff in burned areas. Urgent restoration methods such as hillside sod and seed-spraying treatments in burned areas have been determined to be useful for preventing soil erosion (Ryu et al. 2017). Much of the ^{137}Cs inventory is in the top layers of the soil due to the litter recycling described, and burning of the surface duff layers will affect radionuclide cycling, as well as altering the forest ecosystem. The effects of fire depend on burn severity, with light surface burns at cooler temperatures being much different from severe crown fires. Enhanced leaching of ^{137}Cs will likely occur following a fire, but this leaching may only be to the rooting zone where surviving trees or new plants will access it (Ryu et al. 2017). Planting of trees or shrubs is a long-term option to help fix the contaminants on the site.

Nearby public exposure potential and contamination from fly ash

Responding to the wildfire, the nearby public needs to be alerted in a timely manner and properly informed of protective actions to minimize the potential exposure during a wildfire. Following a wildfire, the impacted community may require remediation to clean up the radioactive PM. This remediation may involve removal of radioactive particles deposited on the surfaces of homes, streets, and other infrastructure. Performing this remediation prior to the first precipitation event will minimize further spread of contamination. If a U.S. city finds itself in the near vicinity of a radionuclide-contaminated forest, a key element of its preparedness should include planning for rapid decontamination of infrastructure and management of the resultant aqueous and solid waste following a wildfire in the contaminated forest.

Conclusion

There is a potential for emissions of radionuclides such as cesium-137 from a wildfire in a radionuclide-contaminated forest. We conducted a laboratory simulation study of a wildfire involving pine needles and peat forest doped with nonradioactive cesium. In this study, emissions of cesium were measured as a function of aerodynamic particle size and compared to cesium content of the residual ash.

Although these experiments were performed on a limited range of biomass types, under a limited range of combustion conditions, using a single chemical form of Cs, they represent the first efforts to assess the potential distribution of radionuclides resulting from a wildfire in a contaminated forest. Future studies could assess exposure to individuals at lower sampling heights, fires involving other potential radionuclides and/or chemical forms of radionuclides, different combustion conditions, or different types of biomass.

In these tests, only 1–2.5% of the cesium on the pine needles burned was emitted as cesium in the airborne particulate matter. Peat fires did not emit cesium into the air. Most of the cesium was concentrated on particle sizes larger than 10 μm.

These data will be used in the future to model potential exposures of airborne particulate containing radionuclides to the surrounding public and firefighters.

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