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# Influence of variable oxygen concentration on the combustion derived release of radiocesium from boreal soil and peat



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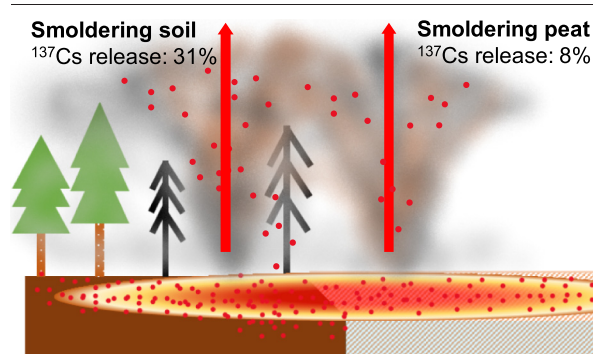
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## HIGHLIGHTS

- <sup>137</sup>Cs emissions from soil and peat were studied in a set of combustion experiments.
- Combustion conditions were varied by altering the oxygen concentration.
- Flaming combustion emitted significantly more <sup>137</sup>Cs than smoldering combustion.
- The relative <sup>137</sup>Cs release from soil was higher than for peat.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Radiocesium, <sup>137</sup>Cs, is one of the most common and dispersed human-made radionuclides. Substantial stocks of <sup>137</sup>Cs are stored in organic layers, like soils and peat, as a consequence of nuclear weapons fallout and accidental releases. As climate warming progresses these organic layers are subject to enhanced risks of wildfires, especially in the vast boreal biome of the northern hemisphere. Reemission of <sup>137</sup>Cs to the atmosphere is therefore presumed to increase. Here, we experimentally investigated the emissions and redistribution of <sup>137</sup>Cs in smoldering fires of boreal soil and peat by varying the oxygen concentration during combustion. For both soil and peat, significantly more <sup>137</sup>Cs was released through flaming combustion in 21% O<sub>2</sub> (50% and 31%, respectively) compared to smoldering in reduced O<sub>2</sub> environments (14% and 8%, respectively). The residual ashes were heavily enriched (>100%) in <sup>137</sup>Cs. Hence, after a wildfire induced volatilization of <sup>137</sup>Cs, there exists further pathways of <sup>137</sup>Cs enriched ash to proliferate in the environment. These results serve as a link between wildfire combustion conditions and the mobility of the <sup>137</sup>Cs inventory found in ground fuels of the boreal environment and can be valuable for radiological risk assessments in a warmer and a more nuclear energy reliant world.

## 1. Introduction

The boreal biome comprises approximately 30% of the global forests (Brandt et al., 2013). The forests and additional macro environments, such as peatbogs, store large amounts of carbon (Bradshaw and

Warkentin, 2015; Pan et al., 2011). The occurrence of wildfires is expected to increase in the northern boreal biome due to climate change (Boulanger et al., 2014; de Groot et al., 2013; Drobyshev et al., 2015; Shvidenko and Schepaschenko, 2013; Stephens et al., 2014). Within the 21st century, the boreal biome is forecasted to experience the most significant increase in

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temperature of all forested biomes (Price et al., 2013). In a scenario of 4 °C global warming, temperature increments of 4–11 °C are expected in the boreal biome. This temperature increase together with an insufficient increase in precipitation are expected to result in a net drying of the boreal biome, hence increasing the probability of wildfires (Flannigan et al., 2016).

Increased utilization of nuclear power is considered as a possible mitigation strategy to limit the CO<sub>2</sub> emissions from fossil fuels and the effects of global warming (IEA, 2021). Approximately 450 civilian nuclear power plant reactors are in operation globally, and 50 more reactors are under construction (Engler, 2020). However, nuclear energy is associated to environmental risks. Globally, eight core meltdowns have occurred in nuclear power plant reactors since the 1950s. History has shown that the frequency of nuclear accidents is higher than first expected, hence the probability of nuclear power plant disasters has been underestimated (Lelieveld et al., 2012; Rose and Sweeting, 2016).

Radiocesium, <sup>137</sup>Cs, is a high-yield fission product from nuclear reactors and is regarded as one of the most hazardous radionuclides released after a nuclear power plant accident. The long half-life (30.2 y), its gamma radiative properties and its high biological uptake by organisms make <sup>137</sup>Cs highly problematic in both the acute as well as in the recovery phase following a nuclear power plant disaster (Endo et al., 2015; UNSCEAR, 1977). <sup>137</sup>Cs has become an omnipresent environmental contaminant as it has been released in substantial amounts as a result of nuclear tests as well as major nuclear accidents (Jones, 2008; Povinec et al., 2013; Steinhäuser et al., 2014).

After ground deposition, <sup>137</sup>Cs is taken up by biological organisms and thus transferred into the biomass of ecosystems (Baklanov and Sorensen, 2001). In a contaminated terrestrial forest ecosystem, <sup>137</sup>Cs will be distributed between the organic soil, ground litter and the vegetation (Pumpanen et al., 2016). The <sup>137</sup>Cs is, even after more than 10 years after fallout, distributed in the top 10 cm layer of boreal soils and peat (Martinsson et al., 2021; Pumpanen et al., 2016; Suomela et al., 1999). Hence, in a wildfire a large fraction of the biologically bound <sup>137</sup>Cs is expected to be re-emitted in the ecosystem, either as ground-based ash or by emissions into the atmosphere by aerosol particles. In a forested environment, high intensity wildfires are denoted crown fires. Crown fires may affect the whole vertical profile in a terrestrial forest, i.e., organic soil layers to the crowns of the trees with a temperature >600 °C (Butler et al., 2004; Certini, 2005; Gustafsson et al., 2019). This will lead to thermal degradation and further ignition of the ambient combustibles. Hence, it is expected that a large fraction of the bio-incorporated <sup>137</sup>Cs will be affected and subsequently re-emitted by the fire. Wildfire induced atmospheric <sup>137</sup>Cs releases have been observed in previous studies (Strode et al., 2012; Wotawa et al., 2006). Experimental investigations have revealed that the biomass bound <sup>137</sup>Cs emitted as aerosols compared to the unburnt fuel has shown to vary between 30 and 70%, and the enrichment of <sup>137</sup>Cs in ash can vary between a factor 2–20 (Amiro et al., 1996; Hao et al., 2018; Martinsson et al., 2021). High temperatures and ample oxygen concentration may lead to conditions with very effective and complete combustion. Such burning conditions commonly result in aerosol emissions that are dominated by inorganic alkali salts, e. g. potassium sulfate (K<sub>2</sub>SO<sub>4</sub>) and potassium chloride (KCl) (Ljung and Nordin, 1997; Nystrom et al., 2017). Hence, it is likely that Cs, which is also an alkali metal, will exhibit similar emission paths as K and consequently also be emitted in high quantities during effective combustion. The most intense combustion condition treatments in the experiments of Amiro et al. (1996) as well as Martinsson et al. (2021) resulted in a release of <sup>137</sup>Cs ≥ 50% of the initial fuel activity.

Smoldering combustion is a low-intensity, low-temperature, and flameless type of combustion of solid fuels (SFPE, 2016). A flaming combustion process can evolve into smoldering by reduction of the oxygen concentration (Wang et al., 2017). Huge carbon stocks that may be subjected to smoldering wildfires are found in the boreal organic soil layer as well as the peat of vast boreal mires (Turetsky et al., 2011; Walker et al., 2019). If dry enough, these environments would support a smoldering combustion rather than flaming due to the limited supply of oxygen to reach these organic layers (Belcher et al., 2010; Sinclair et al., 2020). Once ignited,

these fires can continue to smolder in the organic layers for days to months (Scholten et al., 2021). It has been shown that wildfires affecting peatbogs can combust down to 5 cm of the peat in pristine peatbogs, while for drained or afforested systems the typical combustion depths may reach 10 cm (Wilkinson et al., 2018).

There is currently inadequate knowledge regarding the magnitude of <sup>137</sup>Cs resuspension from smoldering wildfires of soils and peat. Kuzmenkova et al. (2020) investigated the <sup>137</sup>Cs emissions from a burning peatland in the Chernobyl fallout contaminated Bryansk region. The emission factors of <sup>137</sup>Cs from the burned peat was not investigated, although the soil-to-air resuspension factor, *K<sub>r</sub>* (Kashparov et al., 2000), was estimated to  $2.7 \cdot 10^{-6} \text{ m}^{-1}$  based on the activity concentration of the peat and the collected ambient aerosol filters. Although this number is very imprecise since it excludes dilution of the radionuclides in air and thereby obstructs the possibility to extract an emission factor, it suggests a low release of <sup>137</sup>Cs due to the fires.

Here, we experimentally investigate the <sup>137</sup>Cs release from the combustion of boreal organic soil and peat, and the dependence upon variable oxygen concentrations in an attempt to simulate authentic smoldering wildfires to assess radiological exposure routes that have not yet been considered.

## 2. Materials and methods

### 2.1. Forest soil and peat sampling

Forest soil were sampled on the 15th of April 2020 in an approximately 10,000 m<sup>2</sup> pine (*Pinus sylvestris*) dominated forest (N 60.75758, E 17.30341) of south-boreal character outside the city of Gävle in Sweden. The location received a ground deposition of 80–150 kBq m<sup>-2</sup> of <sup>137</sup>Cs during the Chernobyl fallout in 1986 (Byström, 2000). Cores from the trunk of standing trees revealed that the trees were 50–70 years old and were 15–35 years during the Chernobyl fallout. The sampled environment was found in a slope (1–5°) and was partially covered with large rocks of the moraine. The understory vegetation consisted of blueberry (*Vaccinium myrtillus*), lingonberry (*Vaccinium vitis-idaea*), and different mosses. A grid of 6 by 4 (*N* = 24) with 5 m distance in all directions was used for sampling. In each sampling point, a soil sample was obtained by pressing a cylindrical iron soil sampler (H: 7 cm, ø: 7 cm) into the ground surface. All soil samples were stored in 250 ml plastic jars.

Peat was collected on the 16th of April 2020 on the Bengtstjärn peatbog (N 60.75368, E 17.26444). The Bengtstjärn peatbog received deposition of 80–150 kBq m<sup>-2</sup> of <sup>137</sup>Cs during the Chernobyl fallout (Byström, 2000). The peatbog had an approximate area of 5000 m<sup>2</sup> and was located in a small valley of pine forest of south-boreal character. The peatbog vegetation consisted of small trees (pine and birch), tall grasses and reed. A grid of 4 by 6 (*N* = 24) with 5 m distance in all directions was obtained for sampling. A shallow peat sample was taken by pressing a cylindrical soil sampler (H: 7 cm, ø: 7 cm) into the ground in each sampling point. All peat samples were put in 250 ml plastic jars. The samples were then transported to the division of Medical Radiation Physics, ITM, Lund University, and stored in a –18 °C freezer.

### 2.2. Gamma-ray spectrometry of fuel material

The collected soil and peat samples were dried at 70 °C in a drying cabinet. The soil samples contained 48% water and required 4–5 h of drying, while the sampled peat contained >90% water and required 24 h drying. Dried samples were then transferred to 250 ml plastic jars. 24 samples of each sample type were prepared through this procedure prior to gamma-ray spectrometry.

The samples were analyzed by gamma-ray spectrometry using a nitrogen cooled high-purity germanium (Ortec p-type HPGe model GM55-P4) detector with an efficiency of 55%. The detection limit of <sup>137</sup>Cs for a 24-h measurement in this set-up is about 1 Bq kg<sup>-1</sup> dry mass. The measurement time per sample ranged from 4 to 8 h with the condition that full-energy

peak of  $^{137}\text{Cs}$  (at 661 keV) would reach 1000 net counts, which would give a stochastic pulse counting uncertainty of approximately 3%. All activity measurements were decay-corrected to the date of sampling. All samples were then stored at room temperature prior to the combustion experiments.

### 2.3. Experiments

Sixteen dried samples from each fuel material (i.e., forest soil and peat) were selected for the combustion experiments. To reach higher authenticity of the experiments, all samples were conditioned in an 80% relative humidity (RH) chamber (CTS, Model C-40/1000) prior to the experiments. The chosen RH represents the average value observed in Sweden in July during the reference period of 1996–2020 (SMHI, 2020). After treatment in the conditioning chamber the forest soil had on average absorbed 4.3% moisture, while the peat had absorbed 12.8% moisture. An equilibrium moisture content (EMC) of 15% for 80% RH has been observed in wood (Camuffo, 2004). Possible cause of deviating moisture content in forest soil is the higher presence of minerals, sand and gravel which are much less hygroscopic than organic, wood-like, material (Leivo and Rantala, 2003).

The combustion experiments were conducted by using a ISO5660-1 cone calorimeter (NIST, 2018) equipped with a Controlled Atmosphere Attachment, manufactured by Fire Testing Technology. The Controlled Atmosphere Attachment is used for testing materials in oxygen-reduced atmospheres. Prior to each experiment, a sample of dried fuel material was placed in a quadrate tray of aluminum foil,  $10 \times 10$  cm, with the upper side open to be irradiated by the external heat flux from the cone heater. Ceramic wool was used as backing material and was placed underneath the sample for insulation as well as to ensure same level of the sample surface with the mounted upper part of the sample holder to obtain the same distance to the cone heater in the repeated experiments. Hence, the same exposed heat flux to the samples could be achieved. A steel mesh was used to keep the fuel samples inside the sample holder during the heat flux exposure and combustion.

The Controlled Atmosphere Attachment has a mounted heating element which irradiates the sample with a pre-set heat flux, in our case with  $20 \text{ kW m}^{-2}$ , set at the center of the sample surface. The chosen heat flux has been observed to represent the burning conditions of smoldering combustion (Hadden et al., 2014). A thermocouple was used to measure the temperature at the supposed sample surface in the sample holder without fuel with the chosen heat flux. The corresponding temperature for  $20 \text{ kW m}^{-2}$  heat flux (without fuel) was measured to  $346 \text{ }^\circ\text{C}$  ( $\pm 67 \text{ }^\circ\text{C}$ ,

standard deviation, SD), which is within the range of previously observed temperatures in smoldering combustion (Hartford and Frandsen, 1992; Krause and Schmidt, 2000).

The oxygen concentration in the chamber of the Controlled Atmosphere Attachment during combustion was reduced to achieve smoldering. A pre-set mixture of pressurized air and nitrogen gas flow of  $150 \text{ l min}^{-1}$  entered the chamber through a valve. The nitrogen gas was used to dilute the air to achieve a pre-determined oxygen concentration. Five different conditions were evaluated in the experiments where oxygen concentration was set to 21, 17, 13, 9 and 5%. Self-sustained smoldering has been observed between 10 and 21%  $\text{O}_2$  in dried wood, while radiation-aided smoldering could occur between 5 and 7.5%  $\text{O}_2$  (Wang et al., 2017). Triplicate samples were used in each treatment, except for the 5%  $\text{O}_2$  treatment which used quadruplicates. The conditions were randomly assigned to the samples. Tap water was connected to the cooling system of the chamber enabling a continuous cooling of the load cell and other fire exposed parts of the chamber. The experimental setup is depicted in Fig. 1.

Each experiment started by removing the heat shutter, thus irradiating the sample with the incident heat flux. Shortly (5–20 s) after irradiation of the sample surface, flammable gases and white smoke appeared from the sample surface. A spark ignitor was mounted between the sample and the heating element, so that the sample was ignited when there was enough of flammable gases and sufficient oxygen concentration in the Controlled Atmosphere Attachment.

Each combustion cycle in every experiment usually lasted 5–20 min. The sample mass decrease was monitored in real-time during each experiment. The heating element was shut off when the sample mass had decreased to a threshold level based on the residual ashes from near-complete combustion of similar fuels derived from Martinsson et al. (2021). This mass endpoint was 17% ( $\pm 5\%$ ) and 29% ( $\pm 10\%$ ) of the initial mass for peat and forest soil, respectively. The higher mass endpoint for forest soil is explained by a higher content of inorganic refractory material, such as minerals, gravel, and sand. Residual ashes from the experiments were placed in 250 ml plastic jars prior to gamma-ray spectrometry measurements.

The modified combustion efficiency (MCE) was used to monitor the combustion efficiency of each experiment. MCE is derived from the  $\text{CO}_2$  and  $\text{CO}$  in the flue gases and calculated according to Eq. (1). The  $\Delta$  denotes the difference from the background concentration of the respective gas. MCE decreased below unity as  $\text{CO}$  increases during hot and oxygen depleted combustion conditions. It has been demonstrated that the MCE is close to unity for flaming combustion where fine fuels are completely

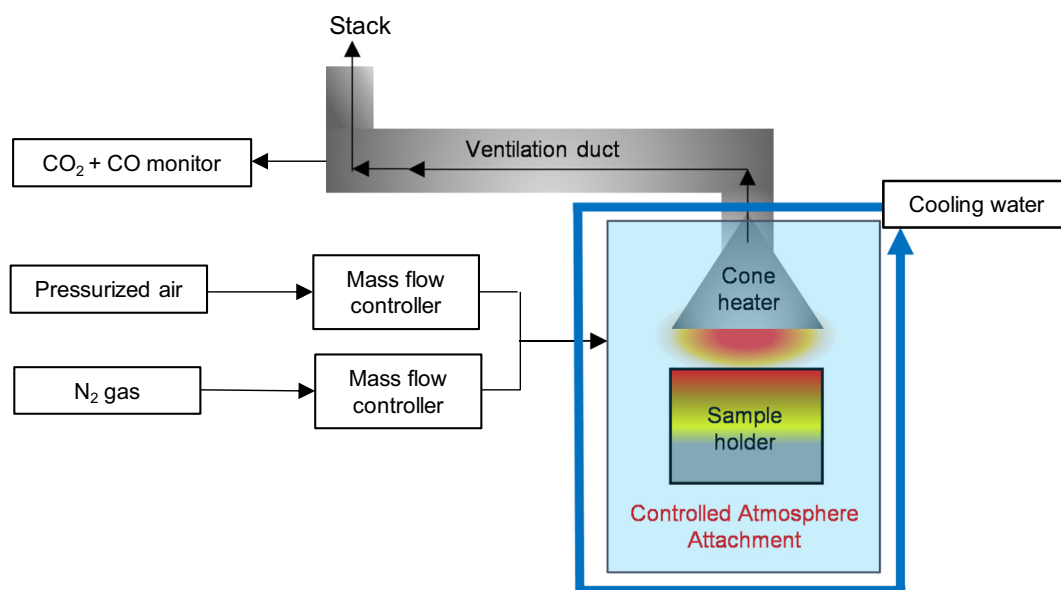


Fig. 1. Schematic of the experimental setup.

combusted (Chen et al., 2007; Yokelson et al., 1996), while MCE values of 0.65–0.85 are common for smoldering combustion (Akagi et al., 2011).

$$MCE = \frac{\Delta CO_2}{\Delta CO_2 + \Delta CO} \quad (1)$$

#### 2.4. Gamma-ray spectrometry of ashes

Gamma-ray spectrometry was used to determine the  $^{137}\text{Cs}$  content in the residual ashes. Since the sample volume of ashes was greatly decreased after the combustion as compared to the initial fuel sample, the sample geometry of the ash samples was altered and consequently the detector efficiency was changed. Adjustments were performed to correct for the altered sample geometry. The filling height of the ashes in the 250 ml canisters was measured. Two standards (NU-651 and NU-652), both containing  $^{137}\text{Cs}$ , of different height (2 and 6 cm) was used to obtain a calibration curve of the detector efficiency as a function of sample height. Hence, a detector efficiency for each sample height could be calculated and used when calculating the  $^{137}\text{Cs}$  activity for each sample. For a single sample, the fractional release of  $^{137}\text{Cs}$  was calculated by comparing the activity of the initial fuel sample to the activity of the residual ashes.

### 3. Results and discussion

#### 3.1. Main characteristics of the fuel and ash samples

The  $^{137}\text{Cs}$  activity concentration of the fuel samples was  $2885 \text{ Bq kg}^{-1}$  ( $\pm 636$ ,  $N = 16$ ) and  $939 \text{ Bq kg}^{-1}$  ( $\pm 397$ ,  $N = 16$ ) for the forest soil and peat, respectively. The  $^{137}\text{Cs}$  activity concentration of the ashes was  $7279 \text{ Bq kg}^{-1}$  ( $\pm 2363$ ,  $N = 16$ ) for the forest soil samples and  $4660 \text{ Bq kg}^{-1}$  ( $\pm 2571$ ,  $N = 16$ ) for the peat samples. The lower activity concentration of the peat fuel samples can be explained by higher mobility of  $^{137}\text{Cs}$  in peatbogs due to much lower abundance of minerals present in the peat, which effectively acts as strong binding sites for the Cs ion (Sato et al., 2013). Hence, by time  $^{137}\text{Cs}$  tends to be more effectively flushed out of the peatbog compared to the forest environment (Stark et al., 2006).

The presence of refractory minerals in the forest soil samples is indicated by the difference in weight between the fuel samples of forest soil and peat, as well as the mass of the ash samples. The forest soil fuel samples had an average mass of  $25.7 \text{ g}$  ( $\pm 19.2$ ,  $N = 16$ ) resulting in a density of  $0.1 \text{ g cm}^{-3}$  while the average mass of the peat fuel samples was  $12.1 \text{ g}$  ( $\pm 5.4$ ,  $N = 16$ ) and a density of  $0.05 \text{ g cm}^{-3}$ . As indicated by the standard deviation of the fuel masses, the soil samples were more heterogeneous than the peat samples. Some of the soil samples contained visibly large fractions of minerals, often in the form of sand, while some samples mainly consisted of black or brown soil together with semi-decomposed litter. This heterogeneity of the forest samples was also manifested in the mass of the ash samples. The average forest soil ash sample mass was  $10.7 \text{ g}$  ( $\pm 17.2$ ,  $N = 16$ ), the peat ash sample mass was  $2.2 \text{ g}$  ( $\pm 0.9$ ,  $N = 16$ ) resulting in a combustion derived mass loss of  $69\%$  ( $\pm 17.3$ ,  $N = 16$ ) for the forest soil and  $81\%$  ( $\pm 6.4$ ,  $N = 16$ ) for the peat. The mass losses also indicate that for most experiments the pre-set mass endpoint was reached, or close to the reference values obtained from near-complete combustion. The mass endpoint was reached in 11 and 13 out of 16 experiments for the forest soil and peat, respectively. The remaining average mass for all experiments was slightly higher (forest soil:  $31\%$ ,  $\pm 17\%$ ; peat:  $19\%$ ,  $\pm 6\%$ ) than in Martinsson et al. (2021) which can be explained by the milder heat flux irradiation as well as the decreased oxygen concentration.

#### 3.2. The effect of reduced $\text{O}_2$ concentration on the combustion derived release of $^{137}\text{Cs}$ and combustion efficiency

Ignition into flaming fires of the samples occurred at all six experiments in  $21\% \text{ O}_2$ , but only in three out of six experiments in  $17\% \text{ O}_2$ , indicating the threshold for smoldering combustion in the  $\text{O}_2$  range of  $17\text{--}21\%$  for

$20 \text{ kW m}^{-2}$ . No flames occurred for the  $5\text{--}13\% \text{ O}_2$  treatments. In Fig. 2, the relative release of  $^{137}\text{Cs}$  during combustion in variable oxygen concentrations are presented. A general tendency of decreased release of  $^{137}\text{Cs}$  with decreased oxygen concentration was observed ( $R^2 = 0.33$ ;  $p < 0.01$ ). The relationship was stronger for peat ( $R^2 = 0.50$ ;  $p < 0.01$ ) than forest soil ( $R^2 = 0.41$ ;  $p < 0.01$ ). Significant differences were observed between the  $21\%$  ( $49.6 \pm 7.5\%$  release) and  $5\%$  treatment ( $14.3 \pm 13.2\%$  release) for forest soil ( $p < 0.05$ ), as well as between the  $21\%$  ( $31.4 \pm 2.1\%$  release) and  $5\text{--}13\%$  treatments ( $7.6 \pm 6.3\%$  release) for peat ( $p < 0.05$ ). Hence,  $3\text{--}4$  times more  $^{137}\text{Cs}$  is released during flaming combustion compared to smoldering combustion in oxygen depleted environments.

The more pronounced effect of reduced  $^{137}\text{Cs}$  release due to decreasing  $\text{O}_2$  concentration in peat ( $R^2 = 0.50$ ) compared to forest soil ( $R^2 = 0.41$ ) might be due to a higher homogeneity of the peat samples. The peat samples were to a much higher extent homogenous near-total organic samples, as indicated by the high combustion derived mass loss ( $81\%$ ) and the low variability in mass loss ( $\pm 6.4\%$ ). On the other hand, and as mentioned earlier, the forest soil displayed a wide range in mass loss of  $19\text{--}86\%$  (mean =  $69\% \pm 17.3\%$ ) due to the various extent of mineral content in the samples. The higher heterogeneity of the soil samples might have induced a higher variability in the  $^{137}\text{Cs}$  release through yet unknown means. The influence of mineral content in smoldering combustion processes of organic ground fuels is not well understood (Reardon et al., 2007). It has been shown that the energy output from smoldering peat decreases with increasing mineral content (Ingram, 1987). Increases in mineral content have also been shown to lower the moisture threshold which would sustain smoldering combustion (Frandsen, 1987). Concurrently, mineral salts of magnesium and calcium, common in soil and plant material, might act as oxidation catalysts and have been observed to increase the propensity of smoldering in wood fibers (McCarter, 1978). Still, combustion of biomass is heterogeneous process and even for a single experimental system the emission properties can vary significantly between the same experimental treatments (Martinsson et al., 2015).

There are inevitable large differences between the resuspension factor,  $K_r$ , and emission factors, such as the  $^{137}\text{Cs}$  release. Still, the relative reduction in  $K_r$  observed by Kashparov et al. (2000) as well as in the  $^{137}\text{Cs}$  release as observed in this study between flaming and smoldering combustion is comparable. Kashparov et al. (2000) conducted field combustion experiments where mainly forest litter was combusted and observed that  $K_r$  was on average 2.8 times higher for the active combustion phase compared to the smoldering phase. Similarly, we observe on average 3.7 times larger  $^{137}\text{Cs}$  release for flaming combustion ( $21\% \text{ O}_2$ ) compared to the

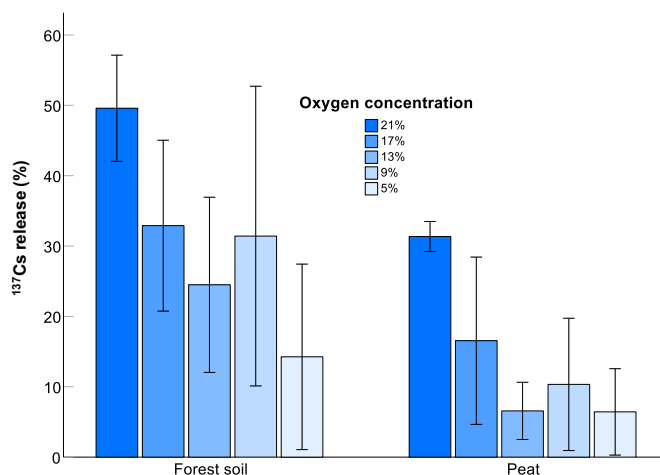


Fig. 2. Mean fractional combustion release of  $^{137}\text{Cs}$  (% released compared to intact fuel) upon exposure of  $20 \text{ kW m}^{-2}$  heat flux in varying oxygen concentrations. Each bar of  $9\text{--}21\% \text{ O}_2$  concentration treatment represents triplicates, while the  $5\% \text{ O}_2$  concentration treatment represents quadruplicates. The uncertainty bars represent one standard deviation of the sample.

smoldering, in the combustion treatments which differs significantly for both soil and peat. Thus, these observations indicate a larger difference in released  $^{137}\text{Cs}$  of ground fuels between smoldering and flaming combustion compared to litter fuels. Still, smoldering appears to be the dominant combustion process of soil and peat in wildfires (Belcher et al., 2010; Sinclair et al., 2020).

When accounting for the different activity concentration of  $^{137}\text{Cs}$  present in ground fuels that are prone to smoldering, and litter layer fuels that are more susceptible to flaming combustion it is possible to calculate an absolute emission factor for the respective fuel for a specific sampled location. The emission factors are calculated by multiplying the activity concentration with the relative release. The observations from this study imply an emission factor of  $403.9 \text{ Bq kg}^{-1}$  ( $2885 \text{ Bq kg}^{-1} \cdot 14\%$  release) and  $75.1 \text{ Bq kg}^{-1}$  ( $939 \text{ Bq kg}^{-1} \cdot 8\%$  release) for smoldering forest soil and peat, respectively. By using activity concentrations and relative release data from Martinsson et al. (2021) the emission factors for flaming combustion of boreal litter layer fuels are obtained for the same sampling location as in this study. The emission factors for forest litter and peatbog litter are estimated to  $130.8 \text{ Bq kg}^{-1}$  ( $545 \text{ Bq kg}^{-1} \cdot 24\%$  release) and  $56.5 \text{ Bq kg}^{-1}$  ( $309 \text{ Bq kg}^{-1} \cdot 18.3\%$  release), respectively. Hence, the emission factors from smoldering peat would be comparable to the flaming combustion of surface forest and peatbog litter. However, for a given ground deposition of  $^{137}\text{Cs}$  ( $\text{Bq m}^{-2}$ ) the smoldering of forest soil would emit a factor 3–7 more  $^{137}\text{Cs}$  ( $403.9 \text{ Bq kg}^{-1}$  vs.  $56.5\text{--}130.8 \text{ Bq kg}^{-1}$ ) due to the significantly higher activity concentration in the soil compared to the other fuels as shown by the emission factors above.

In Fig. 3, the average modified combustion efficiency (MCE) is displayed for each treatment and fuel. For both fuels, the 21%  $\text{O}_2$  treatment had a significantly higher MCE than other treatments ( $p < 0.01$ ), indicating a highly efficient combustion aided by high oxygen supply. The forest soil had a higher average MCE ( $0.94 \pm 0.05$ ) than peat ( $0.90 \pm 0.07$ ) for the 21%  $\text{O}_2$  treatment, although the difference is insignificant ( $p > 0.05$ ). For the 5–17%  $\text{O}_2$  treatments, the MCE ranged from 0.63–0.70 for forest soil and 0.70–0.79 for peat, thereby displaying ranges in MCE which previously have been observed in smoldering combustion (Akagi et al., 2011).

There was a significant correlation between the MCE and the  $^{137}\text{Cs}$  release for forest soil ( $R^2 = 0.30$ ;  $p < 0.05$ ) as well as for peat ( $R^2 = 0.49$ ;  $p < 0.01$ ). Hence, higher combustion efficiency will result in a higher  $^{137}\text{Cs}$  release. These results correspond well with previous observations of increased emissions of inorganic alkali containing ash particles during efficient combustion of biomass (Boman et al., 2004; Johansson et al., 2003;

Lamberg et al., 2011; Leskinen et al., 2014). These studies found mainly enhanced abundance of potassium containing salts, like  $\text{K}_2\text{SO}_4$ ,  $\text{KCl}$  and  $\text{KOH}$ . The chemical similarities of K and Cs, together with increased  $^{137}\text{Cs}$  release during efficient combustion thereby strengthens the potential outcome of Cs to be released in high amounts as well as to form similar salts as K (i.e.  $\text{Cs}_2\text{SO}_4$ ,  $\text{CsCl}$  and  $\text{CsOH}$ ) during efficient combustion.

### 3.3. The effect of reduced $\text{O}_2$ concentration on burn time, mass loss and the enrichment of $^{137}\text{Cs}$ in residual ashes

To reach the pre-set mass endpoint, the samples required sufficient time of exposure of the  $20 \text{ kW m}^{-2}$  heat flux from the cone heater. The average sample required  $673 \text{ s}$  ( $\pm 453 \text{ s}$ ) of irradiation. The forest soil required marginally more exposure time than peat ( $765 \text{ s}$ ,  $\pm 563 \text{ s}$ ;  $587 \text{ s}$ ,  $\pm 313 \text{ s}$ ) but there was no significant difference between the fuels ( $p > 0.05$ ). For both fuels, it required significantly more time to reach mass endpoint for the 5%  $\text{O}_2$  treatment, compared to the 21%  $\text{O}_2$  treatment ( $p < 0.05$ ).

There was a significant difference ( $p < 0.05$ ) in mass loss during combustion between forest soil and peat, where forest soil had a lower mass loss ( $69 \pm 17\%$ ) than peat ( $81 \pm 6\%$ ), which corresponds with previous findings (Martinsson et al., 2021). For both fuels, there was a tendency of reduced mass loss with reduced  $\text{O}_2$  concentration during combustion (Fig. S1). Again, probably due to the presence of refractory minerals, this pattern was less apparent in the forest soil samples than the peat. Hence, there were no significant differences in mass loss between the treatments for forest soil. Mass loss of the 21%  $\text{O}_2$  treatments in peat was significantly higher ( $p < 0.05$ ;  $88 \pm 1.5\%$ ) than the mass loss for the 5 and 9%  $\text{O}_2$  treatments ( $77 \pm 7\%$  and  $76 \pm 5\%$ , respectively).

Mass loss, together with the  $^{137}\text{Cs}$  release have direct effect on the  $^{137}\text{Cs}$  enrichment in ashes. Enrichment was observed for both forest soil and peat, thereby confirming previous field, and experimental observations (Amiro et al., 1996; Martinsson et al., 2021; Paliouris et al., 1995). There was a significant difference ( $p < 0.01$ ) in the enrichment between the fuels, where peat has a factor 2.4 higher enrichment ( $382 \pm 120\%$ ) than forest soil ( $159 \pm 75\%$ ). This ratio is aligned with earlier experimental results (Martinsson et al., 2021), and is the outcome of the higher mass loss of peat compared to forest soil together with the higher  $^{137}\text{Cs}$  release from forest soil compared to peat. Furthermore, for both fuels there was a tendency of reduced enrichment with decreasing  $\text{O}_2$  concentration (Fig. S2), although there were no significant differences between any of the investigated conditions for any of the fuels.

Nutrients present in the boreal ecosystem cycle slowly due to slow decomposition. Hence, like other nutrients, it is probable that the unburned organic matter present in the boreal ecosystem (organic soil, peat and wood) acts as a sink where  $^{137}\text{Cs}$  is stationary for extensive time (Paliouris et al., 1995). As observed in this study, a wildfire will volatilize and resuspend a fraction of the biomass bound  $^{137}\text{Cs}$ . However, as also shown, after the combustion process a majority of the initial  $^{137}\text{Cs}$  present in the fuel will be stored in the enriched ash. This remaining fraction of  $^{137}\text{Cs}$  is likely to be more mobile than before. There are various pathways for continued environmental redistribution of the  $^{137}\text{Cs}$  present in the enriched ashes. The residual ashes themselves may be highly susceptible to slow winds, and previous studies have observed considerable ash resuspension due to wind velocities as low as  $0.4 \text{ m s}^{-1}$  (Etyemezian et al., 2019; Folch et al., 2014; Leadbetter et al., 2012; Liu et al., 2014). Leaching and surface runoff are also two major pathways of continued  $^{137}\text{Cs}$  redistribution. Leaching of  $^{137}\text{Cs}$  will occur to a much larger extent at burned locations than unburned due to increased solubility (Dowdall et al., 2017). Runoff redistribution rates of  $^{137}\text{Cs}$  has also shown to be one order of magnitude higher for burned locations compared to unburned (Johansen et al., 2003; Johansen et al., 2001).

Considering the experimental data available in terms of fuel and ash activity, fuel and ash mass, release and an approximate burn area representative of the sampling location for this study, it is possible to estimate the potential mass and  $^{137}\text{Cs}$  activity of the highly mobile residual ashes which can further disperse and contaminate adjacent areas. The area of

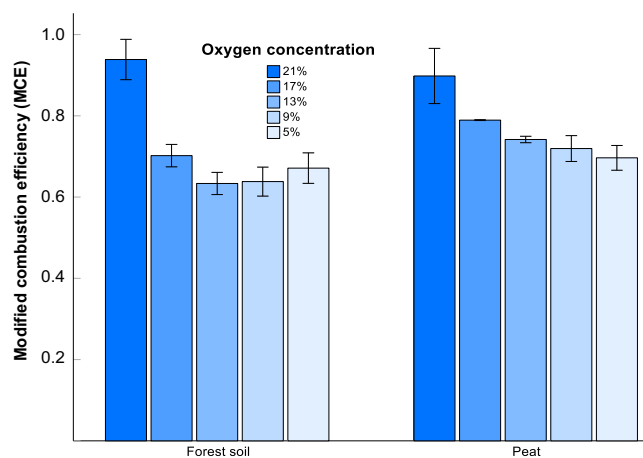


Fig. 3. Mean modified combustion efficiency (MCE) during combustion in the exposure of  $20 \text{ kW m}^{-2}$  heat flux in varying oxygen concentrations. Each bar represents the mean of triplicates in the 9–21%  $\text{O}_2$  concentration treatments, while the 5%  $\text{O}_2$  concentration treatment represents quadruplicates. In turn, each individual data point is an average of a large number of MCE observations ( $N = 200\text{--}2000$ ) due to high time resolution data (1 s) from the cone calorimeter. The uncertainty bars represent the standard deviation.

the sampling locations was approximately 5000 m<sup>2</sup> for the Bengtstjärn peatbog and 10,000 m<sup>2</sup> for the forest site. To facilitate the calculation, we set the area to 1 ha (100 m·100 m = 10,000 m<sup>2</sup>) for both environments. We then use the average mass of the fuel sample for the respective fuel (peat = 0.0121 kg; forest soil = 0.0257 kg), the sampling area of the soil sampler (A = 0.0038 m<sup>2</sup>; H = 0.07 m) and the total area to calculate the hypothetical available fuel mass which is 31.5 · 10<sup>3</sup> kg ha<sup>-1</sup> and 67.7 · 10<sup>3</sup> kg ha<sup>-1</sup> for the peatbog and the forest site, respectively. We can now calculate the total amount of <sup>137</sup>Cs stored in the fuel by multiplying the total fuel mass with the average <sup>137</sup>Cs activity concentration of the fuel (peat = 939 Bq kg<sup>-1</sup>; forest soil = 2880 Bq kg<sup>-1</sup>). Accordingly, the peat would store 29.5 MBq ha<sup>-1</sup> while the forest soil would contain 195.3 MBq ha<sup>-1</sup> of <sup>137</sup>Cs. Now, we apply the average combustion derived release of <sup>137</sup>Cs which is obtained from all experimental treatments (5–21% O<sub>2</sub>) to simulate varying combustion conditions (peat = 14.2%; forest soil = 30.5%). 4.2 MBq ha<sup>-1</sup> and 59.6 MBq ha<sup>-1</sup> would be released by combustion from the peatbog and the forest site, respectively. Finally, by subtracting the released activity from the fuel activity and by applying the mass ratio between the average fuel and ash sample (peat = 5.5; forest soil = 2.4) we estimate the ash activity and mass. The residual ashes from the peat would contain 25.4 MBq ha<sup>-1</sup> of <sup>137</sup>Cs divided on 5781 kg ha<sup>-1</sup> ash while the 1 ha forest soil ashes would retain 135.7 MBq distributed in 28,158 kg ash. The actual ash mass from the forest soil is probably lower due to the content of refractory minerals in both fuel and ash. To conclude, from the results of these calculations it is clear that the amount of <sup>137</sup>Cs that will be spread over a larger spatial scale by combustion and volatilization is far less than what will be left in the highly mobile residual ashes. Hence, the <sup>137</sup>Cs stored in the residual ashes is believed to have a far stronger impact on the local environment compared to the <sup>137</sup>Cs emitted directly through combustion. However, to the best of our knowledge it is still unclear which of the abovementioned transport processes dominates the dispersion of the ash bound <sup>137</sup>Cs.

#### 4. Conclusion

This study demonstrates the controlling influence of combustion conditions on the <sup>137</sup>Cs release from boreal organic ground fuels, susceptible to smoldering wildfires. Flaming and efficient combustion may release 3–4 times more <sup>137</sup>Cs compared to inefficient smoldering. However, when considering the activity concentration of the fuels, smoldering combustion of contaminated soil would emit 3–7 times more <sup>137</sup>Cs compared to smoldering peat and flaming surface litter for the studied area after a wildfire. It is likely that this emission ratio also would be observed in other contaminated heterogeneous boreal environments where the whole vertical fuel profile is affected by fire, implicating that the forested areas should be in focus for fire extinguishing measures to prevent the most extensive spread of <sup>137</sup>Cs. The residual ashes contained a majority (50–90%) of the initial <sup>137</sup>Cs present in the fuel and were heavily enriched in <sup>137</sup>Cs. Hence, the post-fire redistribution of <sup>137</sup>Cs has great potential to proceed through indirect combustion related transport mechanisms such as ash resuspension, leaching and runoff. Future studies should focus on estimation of the activity concentration of size differentiated combustion particles as well as the easily resuspended and enriched ashes derived from flaming and smoldering combustion of <sup>137</sup>Cs contaminated boreal fuels. This knowledge would enable large scale modeling of the atmospheric <sup>137</sup>Cs dispersion due to a forecasted higher wildfire frequency in the boreal ecosystem. Finally, forthcoming investigations should also target the possible complex interactions of extensive soil properties and the combustion derived release of <sup>137</sup>Cs.

#### CRediT authorship contribution statement

Johan Martinsson: Conceptualization, Methodology, Writing – Original Draft, Visualization, Project administration, Funding acquisition.

Guillaume Pédehontaa-Hiaa: Writing – Review and editing, Resources, Validation.

Dan Madsen: Writing – Review and editing, Resources, Validation.

Christopher Rääf: Writing – Review and editing, Resources, Validation, Supervision.

#### Declaration of competing interest

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.

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#### Appendix A. Supplementary data

The supporting information (SI) contains two figures displaying the combustion derived mass loss and the <sup>137</sup>Cs enrichment, respectively. Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.152725>.

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