

## Article

# Influence of Precipitation on the Spatial Distribution of $^{210}\text{Pb}$ , $^7\text{Be}$ , $^{40}\text{K}$ and $^{137}\text{Cs}$ in Moss

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**Abstract:** Mosses have been widely used as biomonitors of a variety of atmospheric pollutants, including radionuclides. Here we determine the radionuclide activity concentration of  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^7\text{Be}$ , and  $^{40}\text{K}$  in moss tissue (*Hylocomium splendens*) collected from 24 sites across Ireland and assess the influence of precipitation on radionuclide spatial distribution. Lead-210 was the most abundant radionuclide (range: 226–968 Bq kg<sup>-1</sup>), followed by  $^7\text{Be}$  (range: <DL–604 Bq kg<sup>-1</sup>),  $^{40}\text{K}$  (range: <DL–155 Bq kg<sup>-1</sup>), and  $^{137}\text{Cs}$  (range: <DL–41 Bq kg<sup>-1</sup>). Albeit nearly thirty years since the Chernobyl disaster,  $^{137}\text{Cs}$  activity concentration was detected at 67% of the study sites; however, the spatial distribution was not fully consistent with the 1986 Chernobyl deposition pattern. Rather,  $^{137}\text{Cs}$  was weakly correlated with rainfall, with higher concentrations along the west coast, suggesting that the 2011 Fukushima Dai-ichi nuclear accident was also a potential source. Average annual rainfall was a significant predictor of  $^{210}\text{Pb}$  activity (linear regression,  $R^2 = 0.63$ ,  $p < 0.001$ ). As such, the highest radionuclide activity was observed for  $^{210}\text{Pb}$  (average: 541 Bq kg<sup>-1</sup>), owing to the high levels of precipitation across the study sites (average: 1585 mm). In contrast,  $^7\text{Be}$  or  $^{40}\text{K}$  were not correlated with precipitation; rather,  $^{40}\text{K}$  and  $^7\text{Be}$  were significantly correlated to each other ( $r_s = 0.7$ ), suggesting that both radionuclides were transferred from the substrate or through soil re-suspension. Precipitation is widely reported as an important factor in the spatial distribution of radionuclides; however, only  $^{210}\text{Pb}$  activity concentrations in moss were strongly influenced by precipitation in the current study.

**Keywords:** biomonitor; ICP Vegetation; *Hylocomium splendens*; activity concentrations; Ireland

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## 1. Introduction

Mosses are widely used as biomonitors of atmospheric deposition because they are broadly distributed (essentially found everywhere), relatively easy to sample, and have a high capacity to trap and accumulate atmospheric particles [1–4]. Moss biomonitoring has been widely used to assess the atmospheric deposition of nitrogen [5–9], trace elements [10–13], persistent organic pollutants [14–17], microplastics [18,19], and radionuclides [20–22]. Numerous studies have shown that radionuclides accumulate in mosses [23–25], making them effective biomonitors for detecting radionuclides that occur in trace atmospheric concentrations.

Since the first nuclear weapons test in 1945, there has been considerable interest in monitoring radionuclides in the atmosphere and their fallout in the environment [26]. Radionuclides are monitored to assess potential human health risks (e.g., the European Commission's Radioactivity Environmental Monitoring program (URL: [rem.jrc.ec.europa.eu](http://rem.jrc.ec.europa.eu)); the Irish Environmental Protection Agency's radiation monitoring program (URL: [www.epa.ie/radiation](http://www.epa.ie/radiation))), quantify atmospheric processes [27,28], predict geological processes, such as erosion [29,30] or sedimentation rates [31], and to gain insight into the paths and processes by which they are transported through the environment [32–34].

Radionuclide activity in moss has been widely reported; nonetheless, studies would benefit from a biomonitoring program to coordinate these efforts similar to heavy metals (i.e., the International Cooperative Programme (ICP) on Vegetation (URL: [icpvegetation.ceh.ac.uk](http://icpvegetation.ceh.ac.uk))).

Radionuclides predominantly come from natural sources, of either cosmic (e.g., beryllium-7 [ $^7\text{Be}$ ]; [35]) or terrestrial (e.g., lead-210 [ $^{210}\text{Pb}$ ]; potassium-40 [ $^{40}\text{K}$ ]) origin. The radionuclide  $^7\text{Be}$  (half-life 53.3 days) is produced by cosmic rays in the lower stratosphere and upper troposphere, where it is adsorbed by particles and ultimately removed from the atmosphere by wet and dry deposition [36,37]. While  $^{210}\text{Pb}$  (half-life 22.3 y) is a progeny of  $^{222}\text{Rn}$  in the  $^{238}\text{U}$  decay series in soils, a fraction of  $^{222}\text{Rn}$  escapes to the atmosphere where it decays to  $^{210}\text{Pb}$ , is adsorbed by particles, and is removed by wet and dry deposition [38]. In contrast, the long-living radionuclide  $^{40}\text{K}$  is found in soil and is only detectable in the atmosphere if soil particles are suspended. Radionuclides also have anthropogenic sources, including medical testing and treatments, nuclear energy plant waste (e.g., Sellafield nuclear power plant waste discharges into the Irish Sea; [39]), nuclear weapons tests (>2000 since 1945), and nuclear power plant accidents. The 1986 Chernobyl and 2011 Fukushima Dai-ichi nuclear power plant accidents widely released caesium-137 ( $^{137}\text{Cs}$ , half-life 30.1 y) into the environment (e.g., 0.085 EBq (exabecquerels) of  $^{137}\text{Cs}$  were released from Chernobyl). Furthermore, commercial processes that use peat have the potential to reintroduce previously deposited (legacy) radionuclides back into the atmosphere [40,41], as is the case with the burning of biomass [23,42]. In Ireland, most of the radiation exposure comes from natural sources, with 55% in the form of radon accumulation in homes. Only 14% of radiation exposure comes from anthropogenic sources, mainly through radiation in medical diagnostics [43]. Nonetheless, radiocaesium was widely dispersed in the Irish environment following the Chernobyl disaster in May 1986, with a mean  $^{137}\text{Cs}$  atmospheric deposition level of 3.2 kBq m<sup>-2</sup> [44]. Climate (notably rainfall) has been highlighted as an important factor in the dispersion and atmospheric deposition of radionuclides [37,45–47].

The objective of this study was to determine the spatial distribution of  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^7\text{Be}$  and  $^{40}\text{K}$  radionuclide activity concentrations in moss (*Hylocomium splendens* (Hedw.) B.S.G.) across Ireland and its relationship to precipitation. All the samples for radionuclide analysis ( $n = 24$ ) were collected under the 2015 ICP Vegetation moss biomonitoring survey [48,49]. We predicted that  $^{210}\text{Pb}$  and  $^7\text{Be}$  would be highly spatially variable and correlated with annual rainfall following the results of previous studies [50–52]. In contrast, we predicted that  $^{137}\text{Cs}$  and  $^{40}\text{K}$  would be less spatially variable and not correlated with annual rainfall, given that potassium and caesium in living moss tissue can be transferred from the substrate on which it grows (e.g., soil) or by lateral transfer between annual growth segments [26]. Radiocaesium concentrations have been measured in *Sphagnum* mosses from Irish blanket bogs [53], but this is the first study to report radionuclide concentrations in *H. splendens* in Ireland.

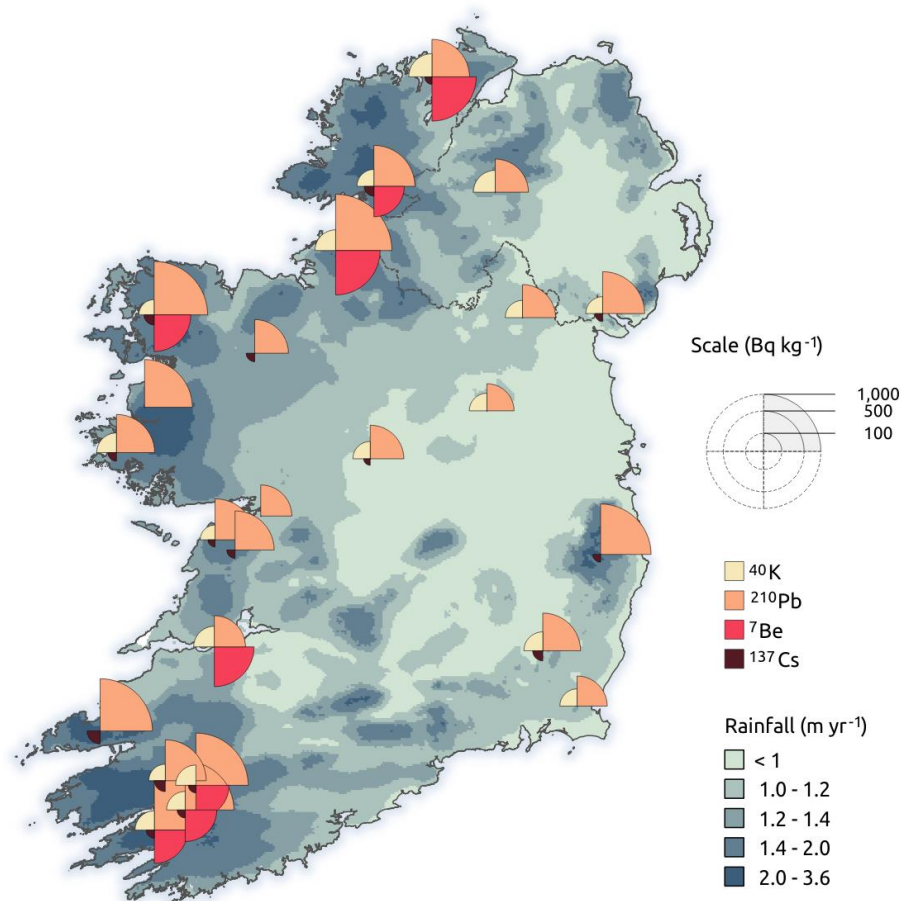
## 2. Materials and Methods

### 2.1. Study Area and Sampling Procedures

Ireland is situated on the northwest periphery of continental Europe; as such, its climate is greatly influenced by the Atlantic Ocean. The prevailing Atlantic winds provide a constant source of relatively ‘clean’ air, as well as marine inputs (e.g., sea salts). The average annual temperatures are moderate, with typically only one day or less below freezing during the winter, and rarely rising above 20 °C during the summer [54]. The average annual precipitation is high, typically consisting of light, but frequent, rainfall. There is generally more rainfall in the west (1000–1400 mm yr<sup>-1</sup>) than the east (750–1000 mm yr<sup>-1</sup>).

During the summer of 2015 (25 May–10 June and 28 July–5 August), *H. splendens* (Hedw.) B.S.G. (see Supplementary Materials Figure S1) moss was collected across Ireland to assess the deposition of trace elements (at 112 sampling locations; for further details on trace elements, see [48]); where the sample mass permitted (>10 g), moss tissue was also

analysed for radionuclides. The subset of samples analysed for radionuclides was predominantly from the west of Ireland, with a few from the central and eastern regions ( $n = 24$ , Figure 1). These moss samples were primarily collected from natural grasslands and heathlands, where the dominant land use was rough grazing, and site elevation ranged from 2 to 265 m above sea level (mean = 121 m). The long-term annual average rainfall across the study sites ( $n = 24$ ) ranged from 979 to 2780 mm (mean = 1585 mm) and the air temperature from 7.9 to 10.5 °C (mean = 9.4 °C [54]). In general, the spatial coverage mirrored that of the wider trace element survey [9,48].



**Figure 1.** Location of sites in Ireland ( $n = 24$ ) where moss (*Hylocomium splendens* (Hedw.) B.S.G.) was collected for radionuclide analysis. Segments represent  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^7\text{Be}$ , and  $^{40}\text{K}$  activity concentration ( $\text{Bq kg}^{-1}$ ); absent segments represent values below the laboratory detection limits. Shading depicts annual average rainfall ( $\text{m yr}^{-1}$ ).

At each sampling location, green moss shoots (representing the last 2 to 3 years of growth; see Supplementary Materials Figure S1) were collected from within an area roughly  $50 \text{ m} \times 50 \text{ m}$  following the ICP Vegetation survey protocols [55]. All samples were collected by hand (wearing nitrile gloves) and composited into paper bags. Every effort was made to gather samples at least 3 m from any tree canopy, 300 m from main roads, and 100 m from smaller roads and houses.

## 2.2. Laboratory Analysis

Moss samples were air-dried at room temperature and any debris or dead material was removed by hand. The samples were then oven-dried at 40 °C to a constant weight and packed into a G100 geometry (cylindrical polypropylene tub filled to 100 mL); the moss sample masses ranged from 10 to 25 g. Radionuclide activity in each sample was

counted in a GMX-15190 n-type coaxial HPGe detector, shielded by 50 mm of lead. Sufficient counting statistics for moss samples requires relatively long measurement periods when using a HPGe detector [36], which limits the number of study sites. Efficiency of the G100 geometry was determined using NPL's mixed radionuclide standard (A150897). Radionuclide activity concentration ( $\text{Bq kg}^{-1}$ ) was determined for  $^{210}\text{Pb}$ ,  $^7\text{Be}$ ,  $^{40}\text{K}$  and  $^{137}\text{Cs}$ ; uncertainty was defined as one standard deviation (see Supplementary Materials Table S1). The measured activity concentrations were corrected for dry weight, and  $^{210}\text{Pb}$  was also corrected for self-attenuation following [56]. Radionuclide analysis was completed within one year of fresh sample collection.

### 2.3. Climate Datasets and Statistical Analysis

Long-term (1981–2010) annual precipitation (Figure 1) and temperature data were obtained from [57]. The distribution of the activity concentration data for each radionuclide was evaluated using the Shapiro–Wilk test for normality. The associations between individual radionuclide activity concentrations in moss, and radionuclide activity concentration and climate (precipitation and temperature) were assessed using Spearman's (non-parametric) rank-order correlation coefficient ( $r_s$ ). Simple linear regression (with variable transformation where needed) was used to determine whether the observed trends were significant ( $p < 0.05$ ). The relationship between climate (and geographic location) and activity concentration for each radionuclide was further explored using multiple linear regression. The spatial variability (%) in activity concentration for each radionuclide across the study sites was estimated using normalized median absolute deviation (NMAD, see Supplementary Materials Equation S1). Lastly, to explore the spatial autocorrelation of radionuclide activity concentration, semivariograms were modelled for each element using the 'gstat' R package [58].

The radionuclide activity concentrations for  $^7\text{Be}$ ,  $^{40}\text{K}$ , and  $^{137}\text{Cs}$  contained left-censored data, i.e., multiple data points were below the detection limit ( $<\text{DL}$ ,  $0.01 \text{ Bq kg}^{-1}$ ). Data imputation was carried out using the 'NADA' (non-detects and data analysis for environmental data) R package [59], which estimates the distribution of values below the censor limit based on the distribution of values above the censor limit. Boxplots were used to display the data distribution of each radionuclide, with values  $< \text{DL}$  estimated through the NADA package. Lognormal transformation of the radionuclide activity concentrations provided a more normal distribution. All statistical analyses were carried out in R 3.3.2 [60].

In general, it is difficult to directly compare radionuclide activity concentrations to published studies, as very few have used *H. splendens* moss. Furthermore, inconsistent methods between surveys can lead to uncertainties [61]; while the ICP Vegetation protocols are widely used and offer consistency, few studies have carried out radionuclide monitoring. In addition, comparison between studies is complicated by confounding factors. It is well established that potassium in moss tissue is either transferred from the substrate (e.g., soil) or by lateral transfer between annual growth segments [26]; therefore, the moss substrate can influence tissue activity concentrations. Furthermore, potassium has an essential physiological function in moss; therefore,  $^{40}\text{K}$  activity concentrations vary throughout the growing season [62]. Radiocaesium activity concentrations in moss reflect residual fallout (deposition) from weapons testing and accidental discharges from nuclear facilities, such as the 1986 Chernobyl accident. Therefore, the distribution of  $^{137}\text{Cs}$  in moss is related to precipitation (washout), meteorology [63], geographical location (e.g., distance from Chernobyl), and time since accidental release into the environment. Furthermore, caesium enters moss tissue via the potassium transport system; therefore,  $^{137}\text{Cs}$  in the environment is expected to follow  $^{40}\text{K}$  to a large extent [20]. This soil-to-plant transfer may influence the interpretation (and comparison) of measured  $^{137}\text{Cs}$  data in mosses [26]. Given the challenges and limitations of inter-study comparison, we cautiously compare our results to published studies, where possible noting the disparate influences of precipitation on the spatial distribution of radionuclide concentrations in moss.

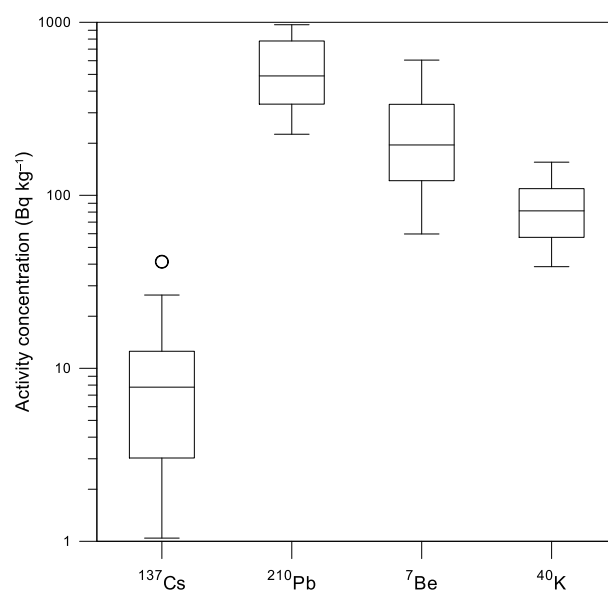
### 3. Results and Discussion

The radionuclide activity concentrations in moss tissue ranged from <DL to 968.04 Bq kg<sup>-1</sup> (fresh weight), with the highest values observed for <sup>210</sup>Pb (Table 1 and Figure 2). Only <sup>210</sup>Pb activity was above the detection limit at all sites (see Supplementary Materials Table S1); the average <sup>210</sup>Pb activity was 541.5 Bq kg<sup>-1</sup> and the median activity was 490.0 Bq kg<sup>-1</sup> (Table 1). In contrast, <sup>7</sup>Be was not detected at >50% of the study sites, likely owing to its short half-life (53.3 days), and the extended period between sample collection and analysis of moss tissue [36]. The observed activity of <sup>7</sup>Be ( $n = 8$ ) ranged from 283.1 to 604.4 Bq kg<sup>-1</sup> (mean = 420.7 Bq kg<sup>-1</sup>, median = 376.2 Bq kg<sup>-1</sup>). Potassium-40 was detected at 75% of the study sites ( $n = 18$ ) and ranged from 57.2 to 155.4 Bq kg<sup>-1</sup> (mean = 96.5 Bq kg<sup>-1</sup>, median = 90.5 Bq kg<sup>-1</sup>). Both <sup>40</sup>K and <sup>7</sup>Be activity concentration data were normally distributed with low spatial variation across the study sites (17 and 22%, respectively; Table 1), potentially suggesting a common source. Furthermore, <sup>40</sup>K and <sup>7</sup>Be were significantly correlated ( $r_s = 0.7$ ; see Supplementary Materials Figure S2), suggesting that both radionuclides were transferred from the substrate or through soil re-suspension. If <sup>7</sup>Be in moss was derived from atmospheric particle deposition, then <sup>7</sup>Be and <sup>40</sup>K would not be correlated [64]. Furthermore, <sup>7</sup>Be was negatively correlated to rainfall across the study sites ( $r_s = -0.45$ ).

**Table 1.** Summary statistics for <sup>210</sup>Pb, <sup>137</sup>Cs, <sup>7</sup>Be and <sup>40</sup>K activity concentration (Bq kg<sup>-1</sup> fresh weight<sup>§</sup>) in moss ( $n = 24$ ). Spatial variability (%) in activity concentration across sites for each radionuclide is represented by NMAD (normalised median absolute deviation).

Statistic	<sup>210</sup> Pb Activity	<sup>137</sup> Cs Activity	<sup>7</sup> Be Activity	<sup>40</sup> K Activity
Non-detect (%)	0	33	67	25
Number > DL	24	16	8	18
Mean	541.5	14.0	420.7	96.5
Median	490.0	10.4	376.2	90.5
Range	225.5–968.0	3.1–41.4	283.1–604.4	57.2–155.4
NMAD (%)	35	31	22	17

<sup>§</sup> Measured radionuclide activity concentrations (Bq kg<sup>-1</sup>) were corrected for dry weight, and <sup>210</sup>Pb activity concentration was also corrected for self-attenuation.



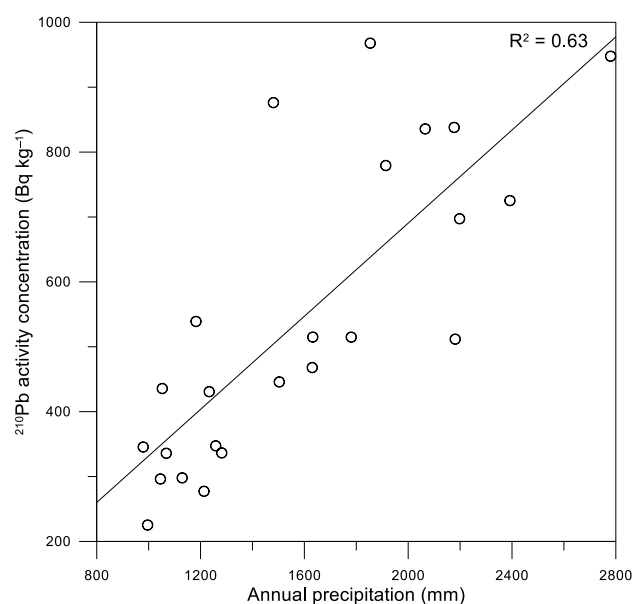
**Figure 2.** Boxplots show the log-transformed activity concentration (Bq kg<sup>-1</sup>) of four radionuclides (<sup>210</sup>Pb, <sup>7</sup>Be, <sup>40</sup>K, and <sup>137</sup>Cs) in moss tissue (*Hylocomium splendens* (Hedw.) B.S.G.) from the 24 study sites in Ireland (see Figure 1). The horizontal line within the box represents the median, box ends represent the upper and lower quartiles, and whiskers represent 1.5 × the interquartile distance. Data imputation for values < DL was carried out using the NADA R package [59].

Few studies have reported  $^7\text{Be}$  or  $^{40}\text{K}$  in moss, and fewer for *H. splendens*. The observed activity of  $^7\text{Be}$  ( $n = 8$ ) was similar to values reported for Serbia (mean = 360 Bq kg $^{-1}$  [64], mean = 314 Bq kg $^{-1}$  [65], mean = 251 Bq kg $^{-1}$  [36]) and Thailand (mean = 226 Bq kg $^{-1}$  [65]). Despite differences in sampling locations, their activity concentrations were similar because  $^7\text{Be}$  is cosmically sourced; as such, its spatial distribution is less affected by processes and events near the earth's surface. Nonetheless,  $^7\text{Be}$  activity concentrations have also been shown to vary by seasons [66]. The observed  $^{40}\text{K}$  activities ( $n = 18$ ) were somewhat lower than the observations reported for Serbia [24,36,67], but direct comparisons are difficult due to differences in the substrate, sampling period, and moss species. As noted,  $^{40}\text{K}$  activity levels in vegetation vary throughout the growing season [62], and  $^{40}\text{K}$  activity concentration in Irish soil has been associated with granite bedrock [68].

The activity concentrations of  $^{210}\text{Pb}$  (225.5–968.0 Bq kg $^{-1}$ ) were comparable to those reported for *H. splendens* (218–913 Bq kg $^{-1}$ ) and *Pleurozium schreberi* (295–1152 Bq kg $^{-1}$ ) from five upland lake catchments in the United Kingdom [69]. In concert with the current study, the catchments were in areas of acid-sensitive geology and elevated rainfall [69]. Similar ranges have been reported in other moss species [25,70]. However, it should be noted that  $^{210}\text{Pb}$  activity between moss species sampled at the same sites can be quite different, in part due to the different physical characteristics of the species [71]. The spatial variability between study sites was the highest for  $^{210}\text{Pb}$  activity concentrations (NMAD = 35%), suggesting high spatial variation in the radionuclide sources; as predicted, the  $^{210}\text{Pb}$  activity concentration was strongly correlated with rainfall ( $r_s = 0.8$ ). Furthermore, the comparison of radionuclide activity with rainfall and temperature indicated a significant positive linear relationship ( $r^2 = 0.63$ ,  $p < 0.001$ ) between rainfall and  $^{210}\text{Pb}$  activity concentration only (Figure 3). Nonetheless, rainfall and temperature were the strongest predictors in multiple linear regression ( $r^2 = 0.76$ , see Supplementary Materials Figure S3), but were only marginally stronger compared with rainfall alone. This suggests that the sites with high  $^{210}\text{Pb}$  activity were primarily the result of higher rainfall volumes. Several studies have reported a positive correlation between precipitation and the atmospheric deposition of radionuclides [47,72,73]; similarly, a decrease in radionuclides in the air has been associated with precipitation [72,74], attributed to 'wash out'. In the United Kingdom, ratios of  $^{210}\text{Pb}$  to Pb in mosses (226–829 Bq mg $^{-1}$ ), soils, and sediments in upland catchments were used to identify sources [69]; the results suggested that the bulk of Pb in moss tissue came from atmospheric deposition, rather than soil re-suspension. Similar ratios of  $^{210}\text{Pb}$  to Pb (155–2518 Bq mg $^{-1}$ ) were found in moss tissues in this study (Supplementary Materials, Table S2), suggesting that the impact of soil re-suspension was negligible, and the  $^{210}\text{Pb}$  activity concentration was dominated by atmospheric deposition.

The observed  $^{137}\text{Cs}$  activities ( $n = 16$ ) ranged from 3.1 to 41.4 Bq kg $^{-1}$  (mean = 14.0 Bq kg $^{-1}$ , median = 10.4 Bq kg $^{-1}$ ; NMAD = 31%). While some studies offer direct species comparison, observed activity concentrations are influenced by location and time, i.e., distance from, and time since, the Chernobyl disaster. The measured  $^{137}\text{Cs}$  activities in *H. splendens* tissue in Italy [75] ranged from 38 to 271 Bq kg $^{-1}$  (mean 121 Bq kg $^{-1}$ ), an order of magnitude higher given the temporal difference (10+ years) with this study. Similarly, the  $^{137}\text{Cs}$  activities in this study were comparatively low when compared with earlier studies that used various moss species in regions closer to Chernobyl (Belarus and Slovakia [20]; Syrian coastal mountains [76]; Serbia [24]). Nonetheless, the  $^{137}\text{Cs}$  activity observed in moss in this study was still related to the Chernobyl accident, given that  $^{137}\text{Cs}$  is prone to lateral transfer [20,26,63]. The  $^{137}\text{Cs}$  activity in moss sampled during 2007 in Belarus reflected the geographic distribution from the 1986 Chernobyl accident [20], suggesting that the measured activity concentrations were not from the recent deposition of  $^{137}\text{Cs}$ , but rather due to the continuous transfer of  $^{137}\text{Cs}$  from soil or from older to younger annual moss segments. However, in the current study, the spatial distribution of  $^{137}\text{Cs}$  in moss was not fully consistent with the 1986 deposition pattern [44]. Rather,  $^{137}\text{Cs}$  was weakly correlated with rainfall ( $r_s = 0.2$ ) with higher activity concentrations along the west coast (Figure 1), suggesting that the  $^{137}\text{Cs}$  activity concentrations in the 2015 moss samples were also

influenced by recent deposition, potentially related to the 2011 nuclear accident at the Fukushima Dai-ichi power plant in Japan. While regional differences in  $^{137}\text{Cs}$  activity concentrations across Ireland were not observed following Fukushima, owing to the significant diffusion of the radioactive plume [77], simulations suggest that cumulated concentrations exceeding  $30 \text{ mBq d m}^{-3}$  could have occurred in western Ireland [78]. This is consistent with the higher  $^{137}\text{Cs}$  activities observed in moss tissue along the west coast (three of the highest activity concentrations: 21.2, 26.5, and  $41.4 \text{ Bq kg}^{-1}$ ), which may reflect the resuspension of material originally deposited to the Atlantic Ocean, as  $^{137}\text{Cs}$  from Fukushima was deposited mostly in the Pacific and Atlantic Oceans [79].



**Figure 3.** Average long-term annual rainfall (mm) versus  $^{210}\text{Pb}$  activity concentration ( $\text{Bq kg}^{-1}$ ) in *Hylocomium splendens* (Hedw.) B.S.G. across the 24 study sites in Ireland ( $R^2 = 0.63$ ,  $p < 0.001$ ).

In Norway, a strong correlation between  $^{137}\text{Cs}$  concentration in *H. splendens* versus deposition rate (correlation coefficient 0.75,  $p < 0.01$ ) was observed immediately following the 1986 Chernobyl nuclear accident, with a ratio of  $1.2 \pm 0.5$  (s.d.) between deposition rate in  $\text{kBq m}^{-2}$  and biomonitor (moss) activity in  $\text{kBq kg}^{-1}$  [22]. During 2011 (after the Fukushima incident in March), the cumulative deposition of  $^{137}\text{Cs}$  in Ireland was estimated at  $10\text{--}50 \text{ Bq m}^{-2}$  [79]; applying the ratio from [22], the expected *H. splendens* tissue concentration after 2011 can be roughly estimated at  $8\text{--}42 \text{ Bq kg}^{-1}$ , which is similar to the observed range in this study (Table 1). While it can be assumed that the majority of the radionuclide deposition from the Fukushima accident to Ireland occurred shortly after the incident (e.g., deposition from the Chernobyl accident occurred in the first 48 h from the time the plume was first detected; [46]), the observed concentration of  $^{137}\text{Cs}$  in green moss tissue generally represents an average of 2 to 3 years of deposition, which is further confounded by the lateral transfer between annual growth segments.

In southern Poland [52], a multiple regression model was used to demonstrate that the deposition of  $^7\text{Be}$  was dependent on the amount of precipitation, but the relationship did not hold true for terrestrial nuclides ( $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ). In contrast, in Serbia, there was no correlation between the precipitation amount or duration and  $^7\text{Be}$  sampled over time [66]. When  $^{210}\text{Pb}$  and  $^7\text{Be}$  activity in mosses was considered as a cumulative measure of deposition and decay, then a strong relationship with precipitation became apparent [80]. We did not observe any correlation between rainfall amount and  $^7\text{Be}$  or  $^{40}\text{K}$ . While  $^{137}\text{Cs}$  deposition was associated with rainfall immediately following the Chernobyl accident [44–46], in the current study,  $^{137}\text{Cs}$  activity in moss tissue and long-term rainfall averages were only very weakly (positively) correlated. Nonetheless,  $^{137}\text{Cs}$  activity in moss tissue



was spatially autocorrelated, as indicated by semivariogram modelling (Figure S4); in contrast,  $^{210}\text{Pb}$  and  $^{40}\text{K}$  showed weak or no indication of a spatial autocorrelation. The spatial clustering of  $^{137}\text{Cs}$  and the general occurrence of higher activity near the southwest coastlines (Figure 1) suggests a relationship with rainfall. The spatial clustering of  $^{137}\text{Cs}$  following Chernobyl was previously identified [46], and when clustering was taken into consideration, a much stronger relationship between daily cumulative rainfall and  $^{137}\text{Cs}$  deposition ( $r = 0.9$ ) was observed in some regions.

#### 4. Conclusions

It is well established that mosses (e.g., *Hylocomium splendens* (Hedw.) B.S.G.) are effective biomonitors of radionuclide activity concentration. Here we found that the  $^{210}\text{Pb}$  activity concentrations had the highest spatial variation across all study sites, and that almost 30 years since the Chernobyl nuclear disaster,  $^{137}\text{Cs}$  activity was detected at 67% of the study sites. Higher  $^{137}\text{Cs}$  activity was associated with higher precipitation regions along the southwestern Atlantic coastline, suggesting that the 2011 Fukushima Dai-ichi nuclear accident was also a potential source. Precipitation volume was a significant predictor of  $^{210}\text{Pb}$  activity concentration in moss (with  $^{210}\text{Pb}$  observed at all study sites). As such, the highest radionuclide activity was observed for  $^{210}\text{Pb}$ , owing to the high levels of precipitation. In contrast,  $^7\text{Be}$  or  $^{40}\text{K}$  were not correlated with precipitation; rather,  $^{40}\text{K}$  and  $^7\text{Be}$  were significantly correlated ( $r_s = 0.7$ ) with each other, suggesting that both radionuclides were transferred from the substrate or through soil re-suspension. Precipitation is widely reported as an important factor in the dispersion and deposition of radionuclides; however, only the  $^{210}\text{Pb}$  activity concentrations in moss were strongly influenced by precipitation in the current study.

**Supplementary Materials:** The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/pollutants3010009/s1>, Table S1: Site ID,  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,  $^7\text{Be}$  and  $^{40}\text{K}$  activity concentration ( $\text{Bq kg}^{-1}$  dry weight) in moss (*Hylocomium splendens* (Hedw.) B.S.G.) tissue ( $n = 24$ ), Table S2: Site ID, location (easting and northing in Irish Grid (m)), elevation (E), precipitation (P), temperature (T), lead-210 ( $^{210}\text{Pb}$ ) activity concentrations and lead (Pb) concentrations in moss tissue, Equation S1: Normalized median absolute deviation (NMAD), Figure S1: Photograph of *Hylocomium splendens* (Hedw.) B.S.G. tissue showing a ‘stair-step’ shape, which is indicative of annual biomass growth. The green shoots are sampled and typically represent the last 2 to 3 years of growth. [Photo credit: Phaedra Cowden], Figure S2: The  $^{40}\text{K}$  activity concentration ( $\text{Bq kg}^{-1}$ ) versus  $^7\text{Be}$  activity concentration ( $\text{Bq kg}^{-1}$ ) in *Hylocomium splendens* (Hedw.) B.S.G. across the study sites in Ireland, Figure S3: Predicted against observed (measured) radionuclide activity concentration ( $\text{Bq kg}^{-1}$ ) for  $^{210}\text{Pb}$  and  $^7\text{Be}$ , Figure S4: Semivariogram for  $^{137}\text{Cs}$  activity concentration ( $\text{Bq kg}^{-1}$ ) in *Hylocomium splendens* (Hedw.) B.S.G. across the study sites.

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