Radionuclides in Mosses

Subjects: Environmental Sciences Contributor: Markus R. Zehringer

The science of the mosses, bryology, knows three main categories of mosses according to constitution and habit. Hornworts (anthocerotophyta), liverworts and bryophytes. The latter are most common. Mosses are non-vascular plants. They adsorb water and nutrients mainly through their leaves. They have a rhizoid only for fixation on the substrate. A water conducting function of the rhizoid is not known. Mosses are suitable as bio-indicator plants. They can take up contaminants and radionuclides directly from the air. Compared to other plants they dispose of a higher surface area. Therefore, there is an ability for accumulation effects. Further, mosses dispose of an active metabolism also in winter. The monitoring of mosses and lichens is well described in literature. Focus in the papers about mosses is on heavy metals, nutrients and air contaminants. Only few papers deal with radio contaminants.

Moss radionuclide fallout radiocesium radiostrontium lead-210

1. Introduction

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2. Collection of the Moss Samples

Within the scope of her school leaving examination, Janine Meyer collected 87 moss samples during the summer 2013 in downtown Basel and surrounding villages. She collected the mosses from top of walls, from tombstones, from rooftops, from trees and on the floor. She considered also mosses grown on trees (epiphytes). Janine removed the mosses from the substrate with help of a scraper or by hand protected with gloves.

2.1. Sample preparation

Each moss sample was categorized by a botanist specialized in determining moss varieties. First, the moss samples were separated from earth particles and stones. Then, they were washed with distilled water. Therefore,

radionuclides adsorbed on the surface of the mosses were excluded. The samples where dried in an oven at 40°C for three hours and weighed (fresh-weight). Finally, Janine dried the moss samples overnight at 120 °C to determine the dry-weight.

2.2. Gamma spectrometry

Janine grinded the dried moss samples and filled them into petri dishes (6.5 cm diameter, fill height 24 mm). The samples were counted with high-resolution germanium detectors for at least 24 hours. Many samples were reanalysed with longer counting times (up to a week) to lower the statistical error (2s=95%) below 10% for the weak emission line of ²¹⁰Pb at 46.5 keV. Janine used the following gamma emission energies (with emission probability in %) for identification and quantification of the radionuclides: ⁷Be: 477.61 keV (10.43), ⁴⁰K: 1460.50 keV (10.67), ¹³⁴Cs: 569 keV (15.4), 605 keV (97.6), 796 keV (85.5), ¹³⁷Cs: 662 keV (84.6), ²¹⁰Pb: 46.5 keV (4.25), ²³⁵U: 143.77 keV (10.94), 163.36 keV (5.08), 185.72 keV (57.0). The gamma spectrometers were efficiency calibrated using a certified ²⁴¹Am/²⁵²Eu-source from the Czech Metrology Institute at Prague. The ²¹⁰Pb peak was calculated using a single point efficiency calculation at 46.5 keV, produce with the measurement of a ²¹⁰Pb solution of 70 Bq/L of the same counting geometry. In addition, a careful correction of the density of the moss samples was necessary (to eliminate attenuation effects).

2.3. Beta spectrometry

Radiostrontium (⁹⁰Sr) was analysed by counting its daughter nuclide, ytttrium-90 (⁹⁰Y) with means of a gasproportional a/b-counter. Both radionuclides are in secular equilibrium, when the sample is older than 20 days. 20-50 g of dried moss were calcinated in an oven at 600°C during four hours. Janine dissolved the ashes in 10% HCI and added yttrium chloride (YCl₃*6H₂O) as a carrier. The she precipitated ⁹⁰Sr and ⁹⁰Y with oxalic acid. The precipitates were calcinated at 850 °C for at least one hour. The ashes were dissolved in 20% HCl and strontium chloride (SrCl₂*6H₂O) added as a carrier. Finally, Janine separated ⁹⁰Y from ⁹⁰Sr by selective precipitation of ⁹⁰Y as yttrium hydroxide. The remaining filtrate, containing the ⁹⁰Sr, was conserved with hydrochloric acid for further trials if necessary. The yttrium hydroxide was dissolved and precipitated as yttrium oxalate. Finally, the oxalate was separated by filtration over a 2.5 cm diameter Millipore-filter. The precipitate was dried at 35 °C and weighed. Finally, the decay of ⁹⁰Y in that source was counted by means of a gas proportional a/b-counter LB 4100 from Mirion during three days (10 consecutive runs of 400 minutes each). The main criterion for the purity of the prepared ⁹⁰Y-oxalate sources was their experimental half-life (theoretically 64 hours) recalculated from the 10 consecutive runs. Almost all moss samples had to be reanalysed twice to thrice due to higher half-lives. The higher half-lives were caused by ²¹⁰Bi present in the source (originated from the high ²¹⁰Pb levels in the moss; half-life of ²¹⁰Bi is 120 hours). After 20 days, ⁹⁰Y in the filtrate of the first precipitation was again in equilibrium with ⁹⁰Sr. The analysis of the further trials led to purer sources. Experimental half-lives of 90 hours or less were acceptable.

3. Results

In the following table, moss data are listed as mean activity values \pm standard deviation of the pooled samples, collected at the same place.

		altitude						
region	substrate	(m) m	⁷ Be	⁴⁰ K	⁹⁰ Sr	¹³⁷ Cs	²¹⁰ Pb	²¹⁰ Pb _{ex}
Hüningen, Kleinhüningen	trees, n=5	250	640 ± 200	1,105 ± 1,690	9.3 ± 11	19 ± 10	450 ± 160	445 ± 165
St. Jakob Park Basel E	trees, n=7	260	455 ± 250	745 ± 515	3.6 ± 2.1	11 ± 7	335 ± 300	330 ± 300
St.Johannspark Basel W	trees, n=3	260	465 ± 270	1,990 ± 2,450	4.5 ± 0.1	11 ± 7	190 ± 120	170 ± 104
Park, Basel E	trees, n=6	260	310 ± 155	1,405 ± 745	1.7 ± 1.9	19 ± 11	390 ± 200	355 ± 190
Lange Erlen Park Basel NE	trees, n=5	260	610 ± 245	675 ± 600	6.0 ± 3.1	15 ± 9.3	400 ± 300	390 ± 225
Park, Basel W	trees, n=3	260	810 ±	390 ± 145	17 ± 15	28 ± 9	605 ± 260	570 ± 255

			320					
Hörnli Graveyard Basel NE	tombstones, n=10	270	840 ± 235	490 ± 280	6.0 ± 8.0	14 ± 10	705 ± 490	695 ± 490
Hardwald, Wood E of Basel	trees, n=2	274	710 ± 70	2,145 ± 710	8.4 ± 0.7	3.7 ± 1	340 ± 140	310 ± 165
Graveyard Basel W	tombstones, n=8	275	485 ± 135	680 ± 540	5.0 ± 4.7	105 ± 100	1,240 ± 565	1,210 ± 565
Rosenfeldpark Basel E	trees, n=2	275	650 ± 50	490 ± 230		6.0 ± 1.3	320 ± 1,510	320 ± 155
SchützenmattparkBasel W	trees, n=4	276	148 ± 34	1,140 ± 1,040	3.7 ± 1.1	17 ± 9.1	415 ± 185	385 ± 155
Sarasinpark Riehen, N of Basel	trees, n=3	280	470 ± 190	355 ± 125	11 ± 12	5.9 ± 0.4	505 ± 240	495 ± 245
Graveyard Wolf, Basel S Basel S	tombstone, n=2	280	675 ± 365	1,335 ± 1,190	56	18 ±10	1,090 ± 390	655 ± 160
Gundeldingen Basel S	trees, walls, n=4	280	640 ± 275	460 ± 100	5.4 ± 3.2	12 ± 4.8	670 ± 500	650 ± 505

Alpweg Village E of Basel	roof, trees, n=5	350	755 ± 560	380 ± 290	9.9 ± 7.2	3.3 ± 2.6	1,989 ± 2,574	1,890 ± 2,580
Bruderholz Park Basel S	trees, n=4	366	545 ± 135	850 ± 595	12 ± 7.6	10 ± 5.8	455 ± 365	450 ± 360
Wartenberg, Hill E of Basel	trees, walls	480	750 ± 185	1,200 ± 1,385	1.7 ± 1.7	21 ±12	1,210 ± 1,045	1,200 ± 935
Tüllingen, Hill N of Basel	tree, n=1	497	170 ± 20	265 ± 45		14 ± 0.8	63 ± 14	51 ± 11
Chrischona, Hill E of Basel	tree, roof, n=2	409	710 ± 410	510 ± 320	10 ± 6.0	28 ± 25	1,520 ± 1,275	1,510 ± 1,290
Gempen, Hill SE of Basel	trees, n=1	760	800 ± 35	1,020 ± 43	7.3 ± 1.5	88 ± 2	467 ± 50	371 ± 40

Table 1 Summarized Moss data. The moss samples were pooled into 20 groups by their find spots and ranged by the altitude of the find spot. All data are in Bq/kg dry weight (dw). n: number of collected moss samples.

3.1. Beryllium-7 (⁷Be)

⁷Be is a cosmogenic radionuclide with a short half-life of 53 days. It reaches the Earth's surface mainly with wet fallout. Only about 10% are deposed dryly.

Due to its short half-live, the presence of ⁷Be indicates that the observed material is recent. In older or dead mosses, ⁷Be would be disintegrated totally.

In all investigated moss samples, ⁷Be was detectable between 150 and 840 Bq/kg dw This shows that the investigated mosses were not older than two years, and no accumulation process in older material did falsify the found activities.

3.2. Found ²¹⁰Pb is mainly unsupported lead from the air

²¹⁰Pb appears at the end of the decay chain of ²³⁸U. It has a relatively long half-live of 22.3 Years. Therefore, it is measurable in soils in activities up to 10 kBq/kg dw. This is the so-called supported part of ²¹⁰Pb, because it is continuously rebuilt in the soil by the disintegration of ²²⁶Ra.

Unsupported or "excess" lead-210 (²¹⁰Pb_{ex})

Part of the built ²²²Rn leaves the soil by Emanation (evaporation of the gas ²²²Rn) into the groundwater and the atmosphere. There, ²²²Rn disintegrates to short-lived daughters and finally to ²¹⁰Pb, which is adsorbed onto dust particles. The disintegrated ²¹⁰Pb cannot be rebuilt from ²²⁶Ra. The latter is no more present (unsupported ²¹⁰Pb). This ²¹⁰Pb is washed out with rain or fallout. The annual fallout flux is estimated to lay between 23 and 367 Bq m⁻² y⁻¹ [1]. Lead is strongly adsorbed onto soil particles of the surface layer of the soils. Mosses and lichens adsorb the ²¹⁰Pb_{ex} directly from the air. The total ²¹⁰Pb-activity is, assuming a secular equilibrium between ²²⁶Ra and ²¹⁰Pb:

Activity (²¹⁰Pb_{tot}) = Activity (²¹⁰Pb_{ex}) + Activity (²²⁶Ra)

Therefore, ²¹⁰Pb_{ex} is calculated by subtraction the activity of ²²⁶Ra from the measured ²¹⁰Pb activity.

Between 90 and 100% of the total ²¹⁰Pb in moss, originate from ²¹⁰Pb_{ex}, as mosses incorporate ²¹⁰Pb directly from the air.

3.3. Fallout parameters Radiostrontium and radiocesium

Most prominent fallout parameters are the long-lived radionuclides radiocesium (137 Cs) and radiostrontium (90 Sr). Even over 30 years after the reactor fire at Chernobyl, these fallout nuclides are present in the environment, even enriched in substrates, such as mosses. The activity range of 90 Sr is 2 – 56 Bq/kg dw and for 137 Cs 4 – 105 Bq/kg dw. The continuous raise of contaminated dust from soils can explain this phenomenon. The mosses incorporate the radionuclides from dust particles adsorbed on the plant surface.



Figure 1. Box plot-distributions of the activities in the collected moss samples (in Bq/kg dw).

Mosses grown on a tombstone at a graveyard in Basel West showed the highest activities of fallout nuclides. The graveyard is situated at the border to France, bordering on open, not built-up fields to the west. This is also the main wind direction in Basel. Mosses (orthotrichum anomalum) on a tombstone showed different activities of fallout nuclides: On the west side of the tombstone, activity was highest: (¹³⁷Cs: 206±11 Bq/kg dw). Whereas, activity was remarkably lower at the foot of the tombstone in East direction (¹³⁷Cs: 49±4 Bq/kg dw).



Figure 2. A tombestone showed mosses with different contamination leve.

It seems that exposition to the main wind direction leads to higher depositions of fallout nuclides on surfaces or their accumulation in mosses. In addition, some find spots on hills show higher activities. However, there is no strict correlation between altitude and found activities. There are other factors influencing the degree of the settled fallout, such as sheltering.



Figure 3. Slight Correlation between ¹³⁷Cs-activity in moss and the altitude of the find spots.





Similar results were observed for 137 Cs in trees ^[2].



Figure 5. Correlation between ¹³⁷Cs-activity in moss and the cardinal points. Numbers in columns are percentage of all moss samples.

¹³⁷Cs-activity in moss samples collected at find spots lying in the principal wind directions in Basel and surroundings, West and East, are significantly higher.

4. Conclusions

Mosses are ideal species for a fallout monitoring. They enrich radionuclides directly from the air. he degree of adsorbed radionuclides into mosses depends strongly on the situation of their find spots. Altitude, degree of sheltering, exposition to main wind direction strongly influence the contamination of mosses.

References

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