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MEASUREMENT OF ENVIRONMENTAL RADIOACTIVITY WITH BASIC COUNTING EQUIPMENT

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In June 1959 the Biology Department of Bowling Green State University received a grant from the Atomic Energy Commission (Grant No. BM-66-59) to purchase equipment for a radiobiology laboratory. The grant was obtained for educational purposes and the proposal specified that the equipment would be devoted primarily to class use. Some of the work reported below, however, was subsidized by a research contract with the Atomic Energy Commission AT(11-1)-536.

The following list of equipment was purchased: three basic decade scalers equipped with lead shielded manual sample changers, halogen-quenched end-window Geiger-Muller tubes and preset timers; one amplifier type decade scaler equipped with a gas flow counting chamber, a cylinder of Q-gas and pressure reducing valve; one organic-quenched end-window G-M Tube; one portable G-M counter; a source kit; and a set of absorbers.

The equipment was supplied with instructions for assembly and operation. It was assembled by members of the biology department and every item was in operation within a few days after its arrival.

In utilizing this equipment we have introduced exercises in radiobiology into existing courses; for example, the general physiology course introduced an exercise in measurement of photosynthesis using C^{14}. The bacteriology course introduced an exercise comparing C^{14} uptake from $NaHCO_3$ solution by yeast cells in presence and absence of glucose. The ecology course introduced an exercise measuring the level of gross beta radioactivity in ash from natural materials. In the general biology course a lecture-demonstration, using the portable G-M counter, was introduced. The research techniques course (graduate students) included laboratory work with these instruments, and a few students registered for special problems utilizing them. During the first 9 months, over 60 students gained experience in the techniques and theory of utilizing radioactive isotopes. This number does not include the students exposed to the lecture-demonstration in general biology. The equipment is installed in a room set aside for this purpose and classes using the room are scheduled in advance to avoid conflicts.

A radiobiology workshop has been scheduled for the summer sessions. This is the only new course that has been added to the curriculum. It provides a study of the fundamentals of radiochemistry, the techniques of measuring radioactivity and of using radioactive isotopes in biological experiments.

Although the instruments purchased were well supplied with instruction manuals, these provided no information as to the efficiency of the instruments for different radioactive elements. The efficiency of an instrument is, of course, profoundly influenced by the shape and thickness of the sample, and the distance of the sample from the window of the G-M tube, but a table of efficiencies for specific substances of given sample thickness and distance from the tube will provide a valuable reference source for comparison of different instruments. In

table 1 such data are provided. Table 1 shows that the beta radiation from potassium, which has an energy of about 1.3 MEV, is detected with an efficiency of about 4 to 7 percent with the halogen-quenched G–M tube, if it is within about 1 cm of the end of the tube, and a sample of 1 g or less is contained in a planchet of 7 cm^2 area. Potassium contains naturally radioactive K^{40} in such concentration that one gram of potassium undergoes 1660 disintegrations per minute ($0.75 \times 10^{-3} \mu c$). It is a most convenient source to use for instrument calibration. It is also revealing for the student to compute the number of K^{40} disintegrations per minute in the adult body knowing that the normal potassium concentration is about 2 g/kg of body weight.

The windowless gas flow counter detects about 33 percent of the K^{40} disintegrations. The weaker C^{14} radiations, however, (energy of about 0.14 MEV; Comar, 1955) are detected by the gas flow counter with an efficiency of only 5 percent. The halogen-quenched end-window tubes measure only 0.4 percent of the radiations emitted by C^{14}. In spite of the low efficiency of the end-window tubes for C^{14}, we have used them successfully for experiments employing this isotope. For example, if 25 ml of pond water are labelled with 0.25 μc of $NaC^{14}O_3$ and exposed to natural light for 6 hr, then filtered through a millipore filter, the activity on the filter will usually register 50 or more counts per minute on an instrument having an efficiency factor of 0.004. A five-minute count of such a filter

TABLE 1

Efficiency of various instruments under specified conditions

Radioactive substance	Type of tube	Distance from tube (cm)	Sample* weight (g)	Efficiency
KCl	Halogen Quenched end window, 1.4 mg/cm^2	0.4	1.02	0.051
"	"	0.4	0.40	0.065
"	"	0.4	0.20	0.070
"	"	1.2	0.87	0.043
"	Gas flow†	windowless	0.57	0.33
C^{14}	Halogen Quenched end window, 1.4 mg/cm^2	0.9	0.001	0.004
"	Gas flow†	windowless	0.001	0.05

*The sample was distributed over an area of about 7 cm^2.

†The gas flow counter (Nuclear Chicago D47) was attached to the amplifier type scaler (Nuclear Chicago 186A), the others were attached to basic scalers (Nuclear Chicago 186).

will measure the activity with a certainty of about 85 percent. This is similar to the reproducibility of duplicate samples; hence, there is no virtue in prolonging the counting time of individual samples.

Before we obtained the equipment listed above, the physics department of Bowling Green State University permitted us to use its basic scaler equipped with a halogen-quenched end-window G–M counter. Karl Schurr, a graduate student in biology, collected specimens of various plants and animals, dried and ashed them, and measured radioactivity of ash samples. Some of the data from his Master's Thesis were used to compile table 2. The observed counts per gram per minute are recorded. The disintegrations per gram per minute were computed using the efficiency factor for K^{40} (0.0038) and the $\mu\mu c$ per gram were computed using the relationship 1 $\mu\mu c = 2.22$ disintegrations per minute. Leaves from terrestrial plants showed the highest activities; leaves from aquatic plants showed

less; and ash from most animal tissues yielded counts hardly distinguishable from the background count of the instrument (13 counts/min).

The high values observed for Sycamore and Willow leaves are higher than the highest average values reported by laboratories using more sensitive instruments. Thus, Nehemias and Whipple (1955) found grass leaves to yield the highest gross beta counts of any organisms they studied. The average concentration observed

TABLE 2

Radioactivity of ash from plants and animals (1957)

Material	counts/g/min	disintegrations/g/min	μμc/g
Terrestrial plants			
Sycamore leaves	114	3000	1350
" fruits	15	390	175
" buds	14	370	166
Mixed wood ashes (fireplace)	14	370	166
Willow leaves	101	2660	1200
Hackberry leaves	57	1500	675
Trembling Aspen leaves	50	1310	592
Aquatic plants			
Potamogeton leaves	27	670	319
Vallisneria leaves	28	726	327
Rhizoclonium	20	526	236
Cladophora	11	290	130
Animal			
Duck liver (Scaup)	19	500	215
Mouse legs	8	210	95
Mollusc flesh	8	210	95
Earthworm	2	53	24
Blue gill	1	26	12
Isopods	0	0	0
Snails	0	0	0

TABLE 3

Radioactivity and potassium content of ash from deciduous tree leaves (1958)

Species	Radioactivity (disintegrations/g/min)	Potassium content (%)
Cottonwood, *Populus deltoides*	1400	4.9
Basswood, *Tilia americana*	625	8.6
White Birch, *Betula papyrifera*	3050	11.4
White Oak, *Quercus alba*	2200	6.9
Sycamore, *Platanus occidentalis*	900	5.3
American Elm, *Ulmus americana*	1800	10.6
Catalpa, *C. bignoniodes*	2250	8.7
Maple, *Acer saccharinum*	3200	8.2
Ash, *Fraxinus americana*	3170	10.0
Average	2066	8.3

in their grass samples was 25 μμc per g wet weight; this amounts to about 250 μμc per g of ash, because 10 g of grass leaves yielded about 1 g of ash. During 1958 we made further analyses of ash from leaves of deciduous trees. Ralph Sechriest, a graduate student, did this work. Because potassium is an important component of plant ash, subsamples of ash were sent to the Brookside Research Lab.,

Inc., New Knoxville, Ohio, for quantitative spectrophometric analysis of potassium content. The observed radioactivity and potassium content of ash of nine species are presented in table 3. The average radioactivity was 2066 disintegrations/g/min and the average potassium content was 8.3 percent. Hence, the observed potassium content could account for only 200 disintegrations/g/min or about 10 percent of the observed radioactivity. A smaller group of samples analyzed for potassium in 1957 yielded values similar to those of table 3.

During the summer and fall of 1959 a broader survey of terrestrial plant leaves was made. Many more species were collected than previously, and collections were made at intervals during the season to learn whether the radioactivity of the ash increased as the season progressed. In table 4 the average levels of radioactivity observed during 1959 are compared with the average values from 1958 and 1957. The samples collected in June yielded a slightly higher average value of radioactivity than the July and September samples, but the differences are not statistically significant. The significant feature of this study was that the level of radioactivity did not increase as the season progressed.

TABLE 4

Average radioactivity of ash from terrestrial plant leaves collected at various times

Time	Fall '57	Fall '58	June '59	July '59	Sept. '59
Gross β disintegrations/g/min	2192	2066	1260	1060	1120
$\mu\mu$c/g	985	930	568	477	505

TABLE 5

Average radioactivity of ash from old cornstalks compared with cornstalks and corn leaves grown during summer of 1959

	Old Cornstalks	1959 Stalks	1959 Leaves
disintegrations/g/min	456	550	1490
$\mu\mu$c/g	205	248	670

Each value is the average of 8 or more samples. The difference between stalk and leaves is statistically significant beyond the 0.01 level. These data were obtained with the gas flow counter having an efficiency of 33 percent for K^{40}. The disintegrations/g/min were computed by multiplying counts/g/min by 3. Samples of about 0.1 g in planchets having an area of 7 cm^2 were used.

The 1959 average values were distinctly lower than those observed in 1958 and 1957, and a sample of the willow ash which had been saved from the 1957 collection when counted in 1959 yielded only 1100 disintegrations/g/min. These data suggest that a considerable fraction of the radioactivity detected in these ash samples represents debris from weapons testing and that the reduced levels in 1959 reflect decay of short-lived components detected in 1957 and 1958. The absence of a detectable increase in level of radioactivity during the 1959 season suggests that very little radioactivity was absorbed by the leaves from rain during that season; however, the summer was unusually dry.

During the winter of 1959–60 a collection of cornstalks and leaves from a field near Bowling Green was studied and compared with a sample of old cornstalks collected from my barn at Put-in-Bay, Ohio. The old cornstalks were present in the barn when I purchased it in 1948; so, they had been stored for more than 11 years. The radioactivity of ash from these samples is presented in table 5.

The 1959 cornstalks revealed a level of radioactivity only 20 percent above that of the old cornstalks. But the 1959 leaves revealed a level about 3 times higher than the stalks. The simplest explanation for the higher radioactivity of these leaves is that they absorbed it from atomic debris in rain that washed over them. If this excess radioactivity were absorbed from the soil it is difficult to understand why it would be deposited preferentially in the leaves, and if the minerals absorbed from soil in 1959 included considerably larger amounts of radioactive atoms than were absorbed by the old stalks, one would expect a larger difference between the old corn stalks and those of 1959.

One additional study was carried out with the ash from cornstalks and leaves: a number of discs were cut from an aluminum sheet, measured, weighed, and their weights per cm^2 computed. These were used as absorbers and the transmission through various thicknesses was measured (Harley and Hallden, 1959). In figure 1 the transmission data for radiation from corn leaf ash are presented. Similar data from a sample of KCl were used to establish the line so labelled. In

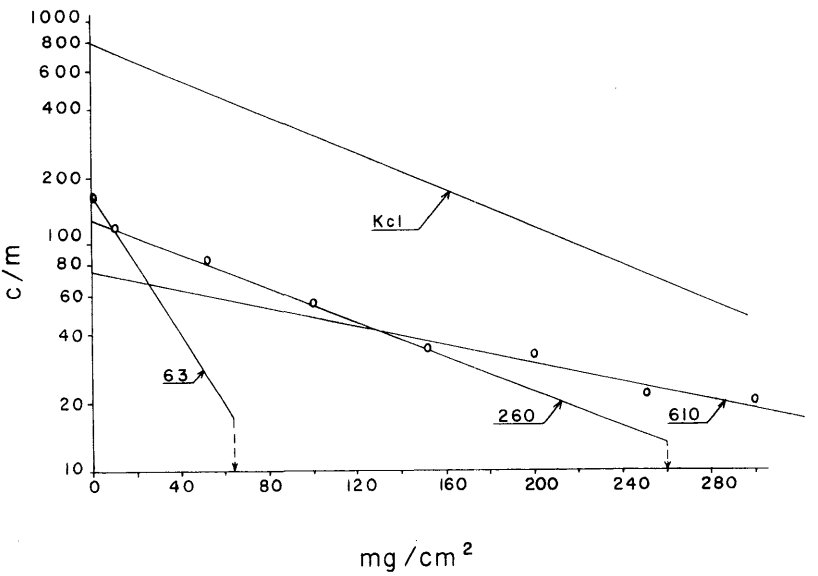

FIGURE 1. Semi-log plot of counts per minute versus absorber thickness (mg/cm^2). Lower plot represents ash from corn leaves collected December 1959. Numbers indicate $t_{0.1}$ values.

analyzing such data it is convenient to draw a line through a set of data and compute the weight per cm^2 required to reduce the transmission to one-tenth of the initial intensity (1 cycle on the logarithmic scale). This parameter, which we will call $t_{0.1}$, is directly proportional to the energy of the beta radiation emitted by the source. Thus, KCl with an energy of about 1.3 MEV has a $t_{0.1}$ value of about 250 mg/cm^2, and C^{14} with an energy of 0.14 MEV has a $t_{0.1}$ value of 28 mg/cm^2. In the ash from corn leaves there seem to be 3 distinct components: a low energy component having a $t_{0.1}$ value of about 63 mg/cm^2, an intermediate component whose $t_{0.1}$ value is closely similar to that of K^{40}, and a high energy component whose $t_{0.1}$ value is about 610 mg/cm^2. Similar analyses of the ash from cornstalks, old and new, revealed only a single component, approximating the line of intermediate slope in figure 1. It seems likely that the low energy component represents a mixture of soft beta emitters. Cerium^{144}, a beta emitter with energies of

0.3, 0.2 MEV and Zirconium^{95} (β, 0.4 MEV) are prominent products of weapons tests (Van Dilla, 1960) and may be important in this portion of the curve. The intermediate slope is certainly influenced by natural K^{40}, but this line also may represent a mixture for there are radioisotopes of energies 1 to 2 MEV in weapons debris which could not be distinguished from K^{40} in this portion of the curve. The high energy portion of the curve may be influenced by Rhodium^{106}, the daughter of Ruthenium^{106}, which is also abundant in weapons debris. Rhodium has a half life of only 30 sec so the disintegration Ru^{106} (β, 0.04 MEV) would be quickly followed by the decay of Rh^{106} (β, 3.5 MEV). The daughter of Cerium^{144}, Praseodymium^{144}, also emits a strong beta (3.0 MEV) and Strontium^{90} (β, 0.5 MEV) has a daughter Yttrium^{90} whose beta energy of 2.2 MEV would place it on this portion of the curve.

An analysis of the kind portrayed in figure 1 will not separate the individual elements in the complex mixture of substances likely to be present in fallout from weapons tests, but it does provide information about the variety of beta energy levels present in the mixture; and the contrast between stalks and leaves suggests that the leaves collect fallout to a much greater extent than do the stalks. The similarity between the radioactivity of ash from old and new cornstalks suggests that most of this radioactivity is contributed by naturally radioactive elements, with K^{40} predominating.

The data presented above demonstrate that radioactivity in ash from natural materials is sufficiently high to be measured with equipment (basic scaler and halogen-quenched end-window G–M tube) now becoming available to many colleges through educational grants from the Atomic Energy Commission. Although measurements of radioactivity in the environment are being made more accurately by laboratories equipped with more sensitive (and expensive) equipment (Nehemias and Whipple, 1960; Setter and Straub, 1958), a student's understanding of the problem is improved by carrying out studies like those described above.

SUMMARY

Instruments purchased with an educational grant from the AEC measured the radioactivity of ash from materials collected in nature. Ash from plant leaves showed levels of about 1000 $\mu\mu$c/g during 1957 and 1958. Leaves collected in 1959 showed levels only one-half so high. Ash from corn stalks was only one-third as radioactive as that from leaves. Surveys of this kind can be carried out by undergraduate students.

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