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Double Activation Analysis: A New Application of Established Techniques

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Large scale environmental tracer options open to ecologists and environmental toxicologists have become limited because few wish to be accused of polluting the very environment they are studying. Neutron activation analysis (NAA) in combination with a stable, nonradioactive element can be an ideal solution to certain large scale environmental tracing and modeling studies (SHUM et al. 1975, JOHANSEN and STEINNS 1976, VAN DALEN and WIJKSTRA 1977, HANSON 1970). NAA-tracer studies utilize elements (activable tracers) which are dispersed into the environment (GREEN 1968a,b). These activable tracers possess properties that allow them to easily be made radioactive upon neutron irradiation (BOWEN and GIBBONS 1963, LENIHAN and THOMSON 1969). When the activable tracer is made radioactive the radioactivity is an accurate measure of the amount of stable tracer in the environmental sample.

A drawback of the NAA-stable tracer technique is that upon introduction to the environment, the stable tracer is "lost" in the medium to be studied until the NAA can be performed. If it is desirable for a study to include the short-term dynamics of a tracer, particularly in aqueous or air systems, sampling therefore becomes a blind exercise until the results of neutron activation are assessed. Short-term dynamics may encompass minutes or hours; the time needed for NAA from collecting the sample to interpreting the data is in terms of one day at best. After such a delay, the time is gone when, if the data warranted it, more samples or samples of a different nature could have been taken. On the other hand, short-term information could be obtained by the use of a radiotracer with a short half-life. But information on long-term dynamics would be impossible to obtain due to decay of the radioisotope.

An ideal tracer would be one that yielded both long- and short-term information, did not contaminate the environment with long-lived radioactivity, and would use the same element in the same chemical form. A study was done using the activable element manganese to determine the feasibility of simultaneously using a short-lived radiotracer in combination with a long-lived stable tracer of the same element in a controlled simulated stream.

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MATERIALS AND METHODS

The artificial stream, which is located out-of-doors, is constructed of wood coated with fiberglass and is 8 m long by 1 m wide by 12 cm deep. Eighteen baffles are spaced alternately along its length to increase the length of water flow to approximately 18 m. Water flowing at a rate of 50 L/min was recirculated in a closed cycle by an electrically driven pump.

Black Willow (Salix nigra Marsh.) shoots were planted in water saturated sod which had become established in the streambed. Five g of Mn as 15.39 g of $MnSO_4 \cdot H_2O$ dissolved in 70 mL of distilled water were exposed to neutron sources containing 20 mg of ^{252}Cf which emit 2.34×10^9 neutrons per sec per mg. If a research nuclear reactor is available as a neutron source, several orders of magnitude less Mn could be used because of the higher neutron flux available.

Stable ^{55}Mn is 100% in isotopic abundance. Neutrons activate stable ^{55}Mn by the reaction, $^{55}Mn(n,\gamma)^{56}Mn$. Manganese-56 has a half-life of 2.58 h and emits gamma rays of 0.847 MeV (99%), 1.811 MeV (29%), and 2.11 MeV (15%). The 0.847 MeV gamma ray was selected to measure both the samples taken after the introduction of the radiotracer into the stream and measurements made during NAA.

A single pulse of 1.85 mCi of ^{56}Mn in 5 g of Mn was added to the stream. On a regular basis for 20 days afterwards samples of water and roots were collected simultaneously. All samples were collected in two dram polyethylene activation vials and counted on a 7.6 x 7.6 cm NaI(Tl) crystal well-detector attached to a multichannel analyzer. After only 12 h the activated ^{56}Mn had decayed to levels below detection. The ^{56}Mn from the original activation used in the immediate tracer study had served its purpose by illustrating the short-term dynamics of the element.

To study long-term dynamics of Mn all samples were taken to the Georgia Institute of Technology to be activated in the Neely Nuclear Research Reactor Center. A portion of the remaining stable Mn which had not been previously activated was activated a second time. The vials containing root samples were flushed with nitrogen gas before activation to reduce the interference from the $^{40}Ar(n,\gamma)^{41}Ar$ reaction. Water samples completely filled the vials. The samples, standards, and background water and root samples containing natural occurring Mn were irradiated in position V-43 at 250 Kw for 2 h at a flux of 4.5×10^{12} n/cm^2-sec. Samples were counted on a multichannel analyzer coupled to a 7% Ge(Li) detector and the data reduced by a programmable calculator. The net photopeak areas of ^{56}Mn were calculated as described previously (COVELL 1959). All values were corrected for radioactive decay and Mn background and divided by the sample volume or dry weight to obtain the counts/mL and counts/g, respectively. From the known quantity of Mn in each standard, the µg quantities of Mn in each sample were calculated. Long, over-night counts of selected root samples made three days after the second activation showed no contribution to the photopeak from long-lived radionuclides.

RESULTS AND CONCLUSIONS

Water samples show a drop from 9.8 µg Mn/mL immediately after ^{55}Mn-^{56}Mn addition to 1.8 µg Mn/mL after 6 h (Figure 1). Root samples show an increase from 750 µg Mn/g soon after introduction of the tracers to 1400 µg Mn/g after 2 h, achieving a maximum of over 4000 µg at 10 h (Figure 2).

The initial activation of stable Mn provided radiomanganese-^{56}Mn for immediate tracer study. The second activation (double activation) activated some of the stable Mn which escaped the initial activation, providing radiomanganese-^{56}Mn for long-term tracer study. The over-lapping data points shown as open and closed circles in Figures 1 and 2 and the continuation of data points for up to 20 days establish the usefulness of the double activation analysis technique for short-term and long-term tracer studies.

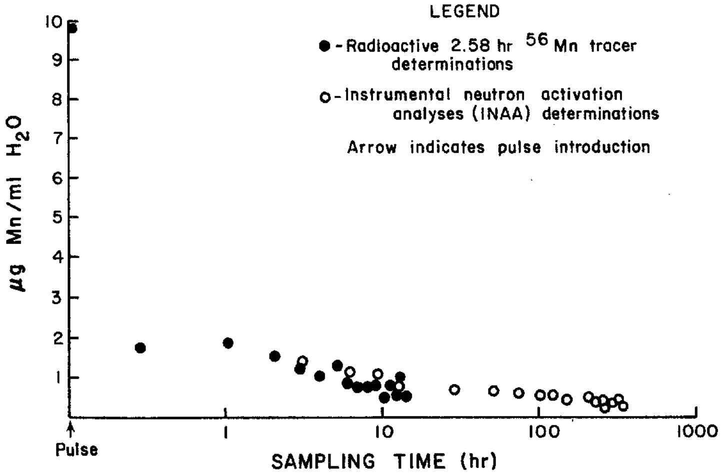

Figure 1. Plot of the µg Mn/mL water versus the log of sampling time.

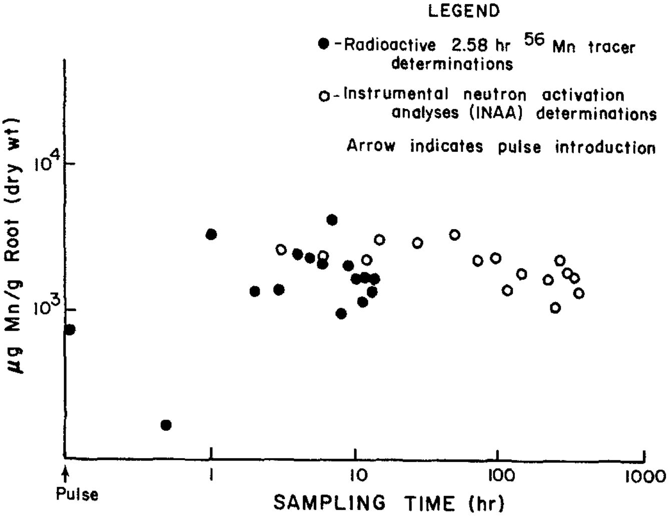

Figure 2. Plot of the log of Mn concentration (μg Mn/g root, dry wt) versus the log of sampling time.

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