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Use of ^{133}Cs and Activation Analysis for

Measurement of Cesium Kinetics in a Montane Lake

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ABSTRACT

The kinetic behavior of cesium was determined in East Twin Lake in the montane zone of the northern Colorado Front Range. The contemplated use of an acute tracer dose of ^{134}Cs was technically feasible but not desirable since the lake is on public lands and used by fishermen. The use of stable ^{133}Cs appeared to have numerous logistic advantages over ^{134}Cs and preliminary studies in the laboratory failed to indicate any major differences in the kinetic behavior of the nuclides at the required concentrations. On September 15, 1970, one kg ^{133}Cs as Cs Cl was uniformly introduced into the $1.45 \times 10^5 \text{ m}^3$ volume of the 5 ha lake. The initial ^{133}Cs concentration in water was about 1400 times less than the $\text{LD}_{50}(5 \text{ d})$ for selected aquatic invertebrates. Samples of water, detritus, seston, vegetation, invertebrates, and trout were removed from the lake at graduated time intervals. Cesium concentrations were measured in approximately 10 g samples by neutron activation analysis. Samples were exposed four hours to a thermal neutron flux of $1.5 \times 10^{12} \text{ cm}^2/\text{sec}$ from a Triga reactor. After a four week decay period, the desired product, ^{134}Cs , could be measured in all samples with a 4 x 8 inch NaI (Tl) crystal-pulse height analyzer system. Preliminary kinetic data on cesium burdens in selected components of East Twin Lake are presented.

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INTRODUCTION

A continuing investigation on the levels of fallout radionuclides in Colorado trout has directed us to intensively study East Twin Lake (ETL) because of the relatively high and persisting ^{137}Cs concentrations in the trout from this lake (Gallegos et al., 1970). Laboratory studies with supporting field measurements have provided various

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kinds of data pertaining to the kinetic behavior of radiocesium in this lake (Gallegos, 1969) but a complete in situ analysis was not possible with fallout ^{137}Cs because the levels in some of the major components were too low for reliable measurement. This paper describes a compartmental kinetics experiment in ETL where stable cesium (^{133}Cs) was used as the tracer. Preliminary findings of this research are presented in support of the feasibility of the stable cesium tracer method.

METHODS AND MATERIALS

The Lake

East Twin Lake is located in the northern Colorado Front Range at an elevation of 2881 meters. Due to its depth and size (Fig. 1), ETL exhibits thermal stratification during the summer months and it may be dimictic, although the spring overturn is usually incomplete.

If estimates of primary production include the aquatic macrophytes, then ETL can be classified as being eutrophic. Vegetation in the littoral zone of ETL (0-2 meters) is abundant and consists of sedge (Carex aquatilis), duckweed (Lemna trisulcata) and water lilly (Nuphar sp.). The most important invertebrates are an amphipod (Gammarus lacustris) and zooplankton of the genera Daphnia and Cyclops. Rainbow trout (Salmo gairdneri) are planted in ETL during July of every year. Trout growth rates (~ 3.5 g/day) are relatively high due to the

abundant food, however, the depletion of oxygen under the winter ice cover usually results in death of the fish before the spring thaw.

Attributes of ETL water such as oxygen concentration, conductivity, temperature and pH were regularly made during the study period. Water conductivity and oxygen content of ETL water four days prior to the labeling event (September 11, 1970) averaged 65 μ mhos and 5.6 ppm, respectively. Water temperatures at this time were uniform with depth and measured 10.7°C. Secchi visibility and pH on the same day were measured as 2.0 meters and 6.2, respectively. Potassium concentrations measured during 1968 in ETL water varied from 1.2 to 1.6 ppm (Gallegos, 1969).

Stable Cesium Feasibility Studies

Initial plans for the cesium labeling experiment in ETL were designed around the use of an acute tracer dose of 0.5 Ci of radioactive ^{134}Cs . Although all aspects of this experiment were feasible, not all were desirable, especially the required closure of ETL to public fishing to prevent humans from becoming contaminated with ^{134}Cs . While the use of stable cesium eliminated the need for closure of ETL to fishing, its use did require certain considerations.

Laboratory experiments were conducted to determine the amount of tracer needed to give detectable ^{133}Cs concentrations in ETL components. This required consideration of the degree of

concentration of ^{133}Cs in a particular component, of the amount of component which could be feasibly sampled and of the sample geometry requirements and irradiation times permitted in the irradiation facility.

Further studies were conducted in the laboratory with Gammarus and Cyclops to determine the acute chemical toxicity of $^{133}\text{Cs Cl}$ to these organisms. Test animals were added to beakers (10/beaker) containing 1.5 liters of ETL water and $^{133}\text{Cs Cl}$ in concentrations varying from 0 to $8.2 \times 10^{-3} \text{ g } ^{133}\text{Cs Cl/ml water}$.

Finally, consideration was given to the fact that the required concentrations of the stable cesium tracer might not exhibit the same kinetic behavior as the originally proposed ^{134}Cs tracer. Beaker experiments were conducted with Gammarus in ETL water to which either $^{133}\text{Cs Cl}$ or $^{134}\text{Cs Cl}$ was introduced to give initial concentrations of 10^{-4} g/l or $1 \mu\text{Ci/l}$, respectively. Two replicate beakers were prepared for each isotope. After 11 days, the concentration factors

Amount tracer cesium/g d.w. Gammarus

Amount tracer cesium/ml water

for stable and radioactive cesium were compared.

Labeling the Lake

On September 15, 1970 one kilogram of ^{133}Cs as $^{133}\text{Cs Cl}$ in solution was metered into ETL by means of electrical pumps from two motor boats which were driven slowly over the entire lake in a systematic pattern. About 15 per cent of the tracer dose was sprayed from one boat into the littoral zone, which comprises 15 per cent of the lake volume while the remaining 85 per cent was pumped from the other boat into the limnetic zone through a perforated hose suspended to a depth of about 3 meters. The entire labeling procedure required one hour to complete.

Collection of Samples

Sample collections were begun 1.5 hours after the labeling event. Collections were made three times during the first day and every twelve hours for the next three days. Daily samples were taken for the next nine days and a biweekly sample for the following three weeks. Currently collections are made at least once a month.

Samples of zooplankton (10-50 grams wet weight) were collected in the limnetic zone with a 12.7 cm diameter Clark-Bumpas plankton sampler towed behind a motor boat. Zooplankton biomass estimates were made by sampling at 30 stations around the lake with a 12.5 cm diameter haul net pulled from the bottom to the surface. This method gave a minimum biomass estimate of 6180 Kg (wet weight) for zooplankton.

During daylight hours on most days, Gammarus were found in the dense mats of duckweed which are confined to the littoral zone. This behavior allowed us to hand sort 10 gram quantities of Gammarus from sweep-net collections of duckweed. The fact that duckweed in ETL grows very near the bottom in the littoral zone and also that Gammarus are almost totally confined to the duckweed plant on bright, clear days, allowed us to make biomass estimates on this invertebrate by systematic sampling of the littoral zone with a 233 cm^2 Ekman dredge. Hand sorting of these dredge samples for Gammarus provided a minimum estimated biomass of 246-Kg (wet weight).

Since the irradiation facility was capable of handling sample containers no larger than 15 cc, a sampling device was constructed for collection of small quantities of sediment. Pieces of conduit were fitted together to form a pole of from 3 to 10 meters long. The lower end of this pole was capable of accepting a 7.5 cm^3 vented polyethylene sample vial. This vial, when fitted into the end of the pole, could be lowered into the sediment to engulf and hold a sample of known area (1.54 cm^2). The small size of the sampling apparatus also reduced the chance of disturbing the sediment around a particular sampling station. Sediment samples were taken at 30 stations around the lake during each sampling period. Total area of bottom sediment was estimated as $6 \times 10^4 \text{ m}^2$ (Gallegos 1969).

Initially water samples were taken at 30 stations (one liter/ station) around the lake using a one liter capacity polyethylene water sampler (Wildlife Supply, Saginaw, Michigan) to determine the degree of homogenous mixing of the ^{133}Cs tracer. However, after the first day post-labeling, one liter samples were taken at both the surface and 0.5 m above the bottom from each of the six stations (Fig. 1). Water samples were filtered by suction through 4.5 cm diameter glass fibre filters (Whatman, GF/A) after collection to remove suspended solids including phytoplankton (collectively called seston).

Samples of sedge, duckweed and waterlilly were obtained from the littoral zone at three locations around the lake. Shears were used to clip sedge and waterlilly at the crown, while a sweep net was used to sample duckweed. Total biomass estimates for sedge and waterlilly were made by clipping and weighing the respective plant materials encountered in a systematically placed quadrat. These estimates were 4953 kilograms and 6080 kilograms of sedge and waterlilly, respectively. Only those sedges growing in ETL water were considered in this estimate. The total biomass estimate for duckweed was made in conjunction with the Gammarus biomass sampling procedure described previously. A value of 9195 kilograms was obtained for this plant.

Fish were taken by hook and line. Length and weight were recorded for each fish and their stomach contents were saved for food habit analysis. A tentative total biomass estimate for trout, based on the number of fish stocked and on the fishing pressure at ETL, was 240 kilograms. More reliable biomass data are being analyzed.

Assay of Samples

All samples were oven or air dried and placed in polyethylene vials (Olympic Plastic Co., Los Angeles, California) which were compatible with the irradiation facility. Standards of known ^{133}Cs concentrations were submitted for irradiation along with the sample materials.

Samples were irradiated for four hours in a thermal neutron flux of about $1.5 \times 10^{12}/\text{cm}^2/\text{sec}$ in the U. S. Geological Survey's TRIGA reactor in Denver, Colorado. Cesium-134 was produced in samples from the capture of a neutron by the ^{133}Cs nucleus (n, γ reaction). Cesium-134 has two gamma rays with energies of 0.6 and 0.8 MeV and a physical half-life of about two years. After a four week period to allow decay of shorter-lived activation products, samples were counted for ^{134}Cs on a 4 x 8 inch NaI (Tl) detector connected to a 400 channel pulse height analyzer calibrated to show 10 keV/channel. Each sample was counted for a length of time sufficient to reduce the standard deviation of the count to within ± 5 per cent of the "true" mean.

RESULTS AND DISCUSSION

Stable Cesium Feasibility Studies

Absolute quantities of ^{133}Cs as small as 6.9×10^{-8} grams were found to be easily detectable after a 12 hour irradiation and gave 900 net counts per minute for the sum of channels 57-63. As a result of this finding we calculated that if we had an initial ^{133}Cs concentration in ETL water of 6.9×10^{-9} grams/ml immediately after labeling, that we would have to collect at least 10 ml samples to detect ^{133}Cs . Laboratory studies indicated that water would determine the minimum amount of tracer dose needed for the cesium kinetics study in ETL since, according to work of Gallegos (1969), this compartment would eventually contain lowest cesium concentrations (a factor of 15 lower than the initial water concentrations). Therefore, the volume of water sampled would have to be increased to about 150 ml to obtain detectable ^{133}Cs concentrations. Since we had originally planned to collect one liter water samples at the various stations, we felt confident that an initial water concentration of 6.9×10^{-9} g ^{133}Cs /ml water would be sufficient to assure measurable concentrations in all lake components. When this ^{133}Cs concentration was multiplied by the volume of ETL (1.45×10^{11} ml), the resulting quantity of tracer ^{133}Cs to be added was 1000 grams.

The chemical toxicity of cesium is poorly known, but the information which does exist suggests that it is relatively low (Cochran et al.

1950, Sax 1963). The acute chemical toxicity of $^{133}\text{Cs Cl}$ to Gammarus and Cyclops as measured in the laboratory by their $\text{LD}_{50(5\text{ d})}$ was about $10^{-5}\text{ g }^{133}\text{Cs Cl/ml}$ water which was about 1400 times the initial tracer concentration expected at ETL ($8.3 \times 10^{-9}\text{ g }^{133}\text{Cs Cl/ml}$ water).

The behavior of ^{133}Cs and radioactive ^{134}Cs in the beaker experiments with Gammarus was very similar as judged by the 11 day concentration factors which averaged 263 and 270 for the respective isotopes.

Sample Gamma Ray Spectra

During the sample irradiation process, numerous activation products other than ^{134}Cs were produced. This presented various degrees of difficulty in resolving the quantities of ^{133}Cs in the sample. The Gammarus spectrum in Figure 2 (curve B, dashed line) was obtained one month after irradiation. Unwanted spectral contributions can be seen at 0.9, 1.1, and 1.3 MeV. The gamma rays producing these photopeaks add to the ^{134}Cs region of the spectrum through Compton scattering processes and, therefore, necessitate the use of spectrum stripping techniques to assess quantities of ^{133}Cs in the sample. However, if given sufficient time for decay, some of these unwanted spectral contributions eliminate themselves from further consideration as shown by the solid lined spectrum for Gammarus (Fig. 2, curve B). The count on this sample was made three months

post-irradiation. The only remaining photopeak at this time, besides the two for ^{134}Cs at 0.6 and 0.8 MeV, was one at 1.1 MeV. This photopeak has been identified as being produced by ^{65}Zn which has a physical half life of 245 days.

Sediment spectra have been the most difficult to resolve and as a result kinetic data for this compartment are not yet available. The sediment spectrum in Figure 2 (curve A) was obtained from a sample which had decayed nearly 2 months after irradiation. There is a photopeak at 0.6 MeV which corresponds to ^{134}Cs , however, two photopeaks at 0.87 and 1.08 MeV tend to dominate the region of interest in the spectrum. The higher energy peaks appear to have physical half lives in the order of several hundred days.

The count spectra for a post-label water (Fig. 2, curve-C) sample including the pre-label background water sample, was obtained one month post-irradiation. The pre-label water sample (dashed line) shows two photopeaks which outwardly appear to be from ^{134}Cs since they peak at energies of 0.57 and 0.77 MeV. However, they are decaying with a physical half-life of a few days compared to the physical half life of 2 years for ^{134}Cs . The occurrence of photopeaks in the ^{134}Cs region of the spectrum which are the result of other radionuclides shows the importance of having a pre-label sample irradiated along with the post-label samples.

Figure 5 shows ^{133}Cs concentrations in some of the components of ETL as a function of time. Detectable quantities of ^{133}Cs were measured in zooplankton and Gammarus samples 1.5 hours and 4 hours after the labeling event, respectively. The concentrations of ^{133}Cs increased rapidly in both organisms and appeared to peak at about two weeks post labeling.

Fish muscle followed the same general pattern but at a much slower rate and at 39 days post labeling, ^{133}Cs concentrations appeared to be still increasing.

Levels of ^{133}Cs in water exhibited a decrease with time and at five days post labeling it was 40 per cent of the initial water concentration.

Concentration factors, based on dry weight of the organisms, were about 3400 for Gammarus and zooplankton (or about 340 based on wet weight). This agrees fairly well with the value of 500 given for zooplankton in studies by Kolehmainen et al. (1968).

The use of stable cesium in a kinetics study such as described has never been documented to the authors knowledge, although the use of stable elements and neutron activation has been used in other areas of biology and medicine (Bethard 1969, Green 1968, and Guinn 1963). Preliminary data suggest that the stable cesium tracer technique as used in ETL appears to be feasible, although the analysis of ^{133}Cs in sediment may prove to be relatively difficult with our

NaI (Tl) detection system. Spectra resolution would be much simpler and more precise with a sensitive Ge (Li) detection system. Experimental data are not available for vegetation but irradiated background vegetation samples appear to contain activation products which are primarily short-lived.

A distinct advantage of the stable cesium technique for systems experiments in situ is that the radiation safety aspects are obviated, thereby allowing public access to the study area. This approach may be useful for studies on lakes or other systems in populated areas where the use of a radioactive tracer may not be desirable.

The thermal neutron activation of sample materials from ETL may result in a simple technique for quantitatively determining the naturally occurring levels of stable elements such as Zn, Cs, Fe, Cl, and Na.

The usefulness of other stable tracers in aquatic studies will depend upon several considerations such as chemical toxicity, quantities of tracer needed and activation properties of the tracer.

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LONGITUDE $40^{\circ}37'$

LATITUDE $105^{\circ}50'$

VOLUME = $145,492 \text{ M}^3$

AREA = $49,936 \text{ M}^2$

MEAN DEPTH = 2.91 M

⊗ = WATER SAMPLES