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4.3 ECOLOGY OF TOXIC METALS

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4.3.1 Abstract

The fate of toxic metals released into the environment is one of the nation's important ecological problems. This report describes experiments on the movement and concentration of mercury and cadmium in different ecosystems. Radiotracer techniques and stable chemical analyses were used in cycling and food chain studies in aquatic and terrestrial ecosystems. Direct uptake and transfer efficiencies through a simple aquatic food chain were determined for $CH_3{}^{203}HgCl$, $^{203}Hg(NO_3)_2$ and $^{109}Cd(NO_3)_2$. Methylmercury was much more efficiently transferred through the food chain than inorganic mercury or cadmium. A linear eight compartment model was developed to describe the dynamic redistribution of methylmercury in a pond ecosystem using data collected by radiotracer techniques. The general mobility of mercury and cadmium in terrestrial ecosystems and the extent to which they concentrate in various components were compared. In the systems tested, data indicated a greater mobility for mercury and the tendency for cadmium to be accumulated in organic components. The transfer of mercury from the terrestrial to the aquatic system was studied in microcosm experiments. Approximately one-half of the radioactive mercury on fly ash which had been applied to the terrestrial component was transferred to the aquatic component of the microcosm after 4 1/2

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Human Subjects Project

The biomagnification of mercury has become a common generalization which is not easily substantiated when the evidence is closely examined. Studies of the aquatic ecosystem and food chain pathways leading directly to man are still important areas of research; however, since a significant quantity of heavy metals are released to the terrestrial ecosystem, the transfer of mercury and cadmium from the terrestrial to the aquatic ecosystem and the cycling of these materials in terrestrial systems are also important.

Our studies[1] on the terrestrial cycling of mercury and cadmium are nearing completion and will be reported in greater detail in this report. The terrestrial studies emphasized the mobility of cadmium and mercury in different systems and demonstrated the transfer of mercury from water to the fruit of plants. Other studies have emphasized land-water interaction or the transfer of toxic metals from the terrestrial to the aquatic ecosystem. Analyses for mercury in fly ash collected on the ORNL Walker Branch Watershed have led to microcosm studies which offer an explanation of concentrations of mercury in fish detected in streams of the area.

Since methylmercury concentrations in fish have not been adequately explained, food chain transfers and uptake and retention studies are increasingly important. Our previously reported aquatic studies involved the efficiency of food chain transfer of different forms of mercury and cadmium. These studies have continued and will be briefly summarized along with the inclusion of new data. Stream tag studies with methylmercury and inorganic mercury were previously reported. Analyses of data from these studies have continued and additional data have been collected on a stream tagged with cadmium.

Information obtained in ecological experiments on the fate of toxic metal released into the environment is being used in the development of exchange submodels in the Unified Transport Model for toxic materials (see section 4.1). Using data collected in a field study an eight compartment linear model has been developed to describe the dynamic redistribution of methylmercury in a pond ecosystem. More detailed analyses of both the data and model are currently in progress.

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compounds were run to check for mercury losses during sample preparation; losses were negligible. The cadmium analyses were by spark source mass spectrometry (SSMS) with isotope dilution.

The mosses (Dicranum and Polytrichum) had higher concentrations of cadmium and mercury than the other vegetation with the exception of mercury in the grass Andropogon (Table 4.3.1). Background concentrations of metals in plants will vary widely because of the complexity of factors controlling metal ion uptake: soil concentration of metals, competing soil cations, species differences in uptake rate, and atmospheric input of metals.

Table 4.3.1. Mercury and cadmium content in vegetation from Cades Cove, Great Smoky Mountains National Park, Tennessee, collected in March 1972

Mercury, ppm air-dried weight; cadmium, ppm oven-dried weight

	Dicranum		*Polytrichum*		*Festuca*		*Andropogon*		*Tsuga*			
									Needles		Stems	
	Hg	Cd	Hg	Cd	Hg	Cd	Hg	Cd	Hg	Cd	Hg	Cd
n	14	14	12	12	6	6	6	6	6	6	6	6
M	0.118	0.421	0.119	0.383	0.087	0.279	0.120	[illegible]	0.027	0.083	0.028	0.326
SD	0.060	0.143	0.046	0.114	0.048	0.500	0.063	[illegible]	0.004	0.052	0.010	0.181

4.3.3.2 Fly Ash as a Source of Mercury Pollution

The discovery of high concentration of mercury in organisms remote from known mercury sources led to identification of fossil fuel burning as a source of regional mercury contamination.[6,7] The average mercury concentration in coal varies so much (< 0.01 - > 100 ppm) that it is unrealistic to cite an average value. Mercury concentrations in coal fly ash collected in areas distant from the discharge stacks by Cannon and Anderson[8] were enriched by a factor of two over bottom ash samples, and fly ash collected from stack precipitators has been shown to contain 0.2 ppm mercury while the coal contained 0.3 ppm mercury.[9] It has been calculated that 90% of the mercury in the coal is lost out of the stack to the environment during normal furnace operations.[9]

Mercury in coal is converted to the vapor phase in the furnace when the coal is burned, but some of it may condense on fly ash

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Cove D. scoparium did not contain significantly more mercury than that found at the highest elevation in the Smoky Mountains. Cades Cove is surrounded by mountains and fly ash originating in the Appalachian Valley would have to pass over a minimum elevation of 600 m before reaching the Cove.

The moss Polytrichum commune was not found at ORNL, but at Cades Cove and higher in the Smoky Mountains it contained less mercury than D. scoparium. The difference in the mercury content of these two species could be due to their growth forms. Dicranum scoparium forms mats, while P. commune grows in more upright and less dense stands which apparently entrap fewer fallout particles.

The mercury concentration in the composite fescue grass Festuca eliator sample at ORNL was 0.025 ppm, compared with the mean value of 1.13 ppm for the D. scoparium. Moss-fescue mercury concentrations were compared at another location, several kilometers from the nearest smokestack. Dicranum scoparium did not grow in this habitat, but the smaller moss species Eurhynchium hians, Brachythecium rivulare, and Sharpiella striatella were found abundantly growing as intertwined mats under heavy grass (Festuca) cover. A complete sample of these mosses contained a mercury concentration of 0.044 ppm, while the overstory grass (Festuca) contained 0.009 ppm mercury. Forbs from the same location were quite variable in mercury concentration but were always at least a factor of 2 less than the mosses.

Eastern hemlock (Tsuga canadensis) needles and stems and Rhodendron maximum leaves collected in the same location as the mosses in Cades Cove contained 0.025 and 0.024 ppm mercury, respectively (Table 4.3.1). Mercury concentrations in these species were significantly lower ($P \leq 0.01$) than the moss concentrations, which indicated that D. scoparium and P. commune accumulate mercury from the environment to a greater extent than the vascular plants examined.

Fly Ash Microcosm Experiment: Since atmospheric derived mercury may be available to the aquatic biota, a microcosm experiment was run to identify target organisms and sinks and to determine the transfer rate of fly ash-rain water derived mercury from the terrestrial to the aquatic ecosystem. (See Fig. 4.3.1.) Duplicate tan-

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(0.3 x 0.3 x 1.6 m) were established and kept at the proper photoperiod (long-day) and temperature (65-70°F) for the duration of the experiment. Sections of a stream bank were placed in one end of the tanks, with stream sediments and 60-ℓ of water in the other half. Fish (Gambusia affinis) and snails (Goniobasis clavaeformis) were placed in the aquatic portion. After the two systems stabilized, radioactive ^{203}Hg-tagged fly ash was added in simulated rainfall. The fly ash tagging was accomplished by placing a coil of copper wire in a solution of $^{203}Hg(NO_3)_2$ overnight, removing and placing the wire over a few grams of fly ash in a flask and heating the wire to incandescence. The ^{203}Hg that had amalgamated onto the wire vaporized off and deposited on the fly ash particles. The tagged fly ash was then mixed with unlabeled fly ash to bring it to the proper activity (~200 μCi/g) and added to the terrestrial portion of each microcosm. Each microcosm received about 110 μCi of ^{203}Hg.

Sampling and counting for ^{203}Hg activity was then conducted on a routine basis for 4 1/2 months. The terrestrial portion of the microcosms was sampled immediately post-tag, then once weekly for five weeks, and once monthly for the duration of the experiment. Terrestrial sampling included litter and soil. Water samples and fish were counted at 24-hr post-tag, once weekly for five weeks, and then once monthly until the experiment was terminated. Snails and sediments were counted once weekly starting at one week and two weeks, respectively, until the fifth week, and then once monthly for the duration of the experiment. The distribution of ^{203}Hg activity at the termination of the experiment (4 1/2 months) is shown in Table 4.3.2. The mercury on fly ash was shown to be leachable and thus available to biota since radioactivity was found throughout the soil column and in the plant roots. Soil invertebrates could have distributed the particles of fly ash down into the soil to this extent but plants that sprouted several weeks after the tag contained ^{203}Hg in the vegetative parts, indicating leaching of the mercury off of the fly ash.

The activity in the sediments could be due to the washoff of fly ash particles, but the activity of the aquatic biota seems more reasonably to be due to the uptake of ionic mercury, rather than to

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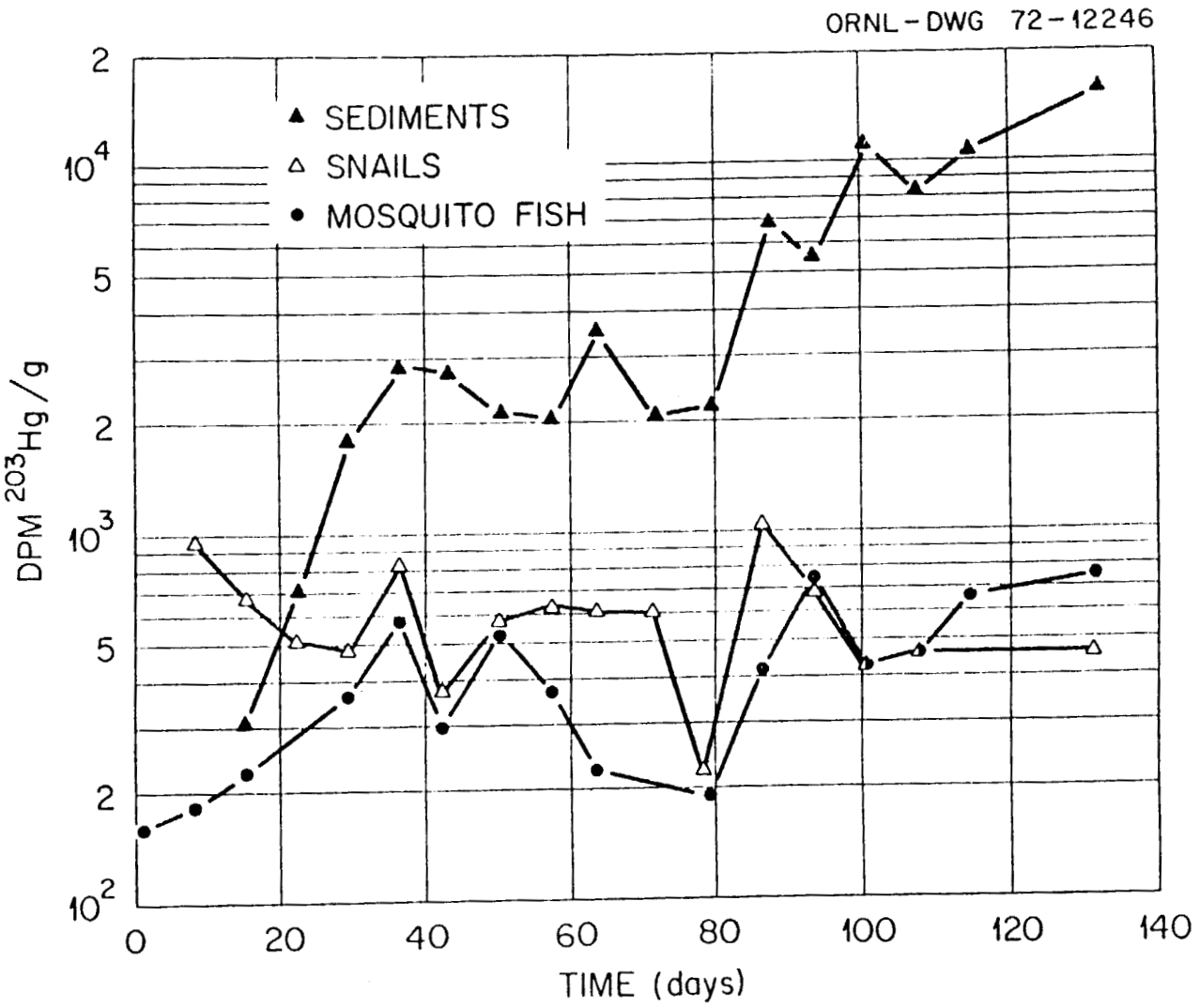

Fig. 4.3.2. Transfer of ^{203}Hg from terrestrial to aquatic portion of a stream bank microcosm tagged with ^{203}Hg labeled fly ash. Snails represent a composite sample. Fish values are averaged for observations, and sediment values are means of six observations.

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4.3.3.3 Cycling of Mercury and Cadmium in Terrestrial Ecosystems

The relative movements of inorganic forms of mercury and cadmium were studied in two terrestrial ecosystems - a small eastern red cedar stand and an old field. The purpose of these studies was to compare the general mobility of these elements and the extent to which they may concentrate in various ecosystem components.

Solutions of $^{203}Hg(NO_3)_2$ and $^{109}CdCl_2$ were inoculated into trunks of eastern red cedar trees and subsequent movements in the trees and transfers via leaching by rain to understory, litter and soil were determined. Table 4.3.3 illustrates the distribution of ^{203}Hg and ^{109}Cd eight weeks after inoculation. While ^{109}Cd was translocated more readily than ^{203}Hg, mercury was more mobile in terms of foliar leaching in rain and transfers through litter to soil. Cadmium translocated into branches and foliage in quantities directly proportional to the weights (determined by the tree harvests) of these plant compartments. Mercury was much less mobile with 80% of the inoculum remaining within several centimeters of the input point. Although only 1.5% of the mercury translocated into foliage, compared to 28.5% of the cadmium, leaching losses of mercury were three times greater than those of cadmium.

Table 4.3.3. Distribution of ^{109}Cd and ^{203}Hg from inoculation into trunks of Eastern red cedar trees eight weeks after inoculation in eastern Tennessee, summer 1972

	^{109}Cd	^{203}Hg
Percent of input		
In trunk	34.6	96.5
In branches	36.8	1.7
In foliage	28.5	1.5
Leached out by rain (20.8 cm)	0.1	0.3
Percent of activity rained out		
In understory	2.7	1.4
In litter	68.0	16.0
In soil	29.3	82.6

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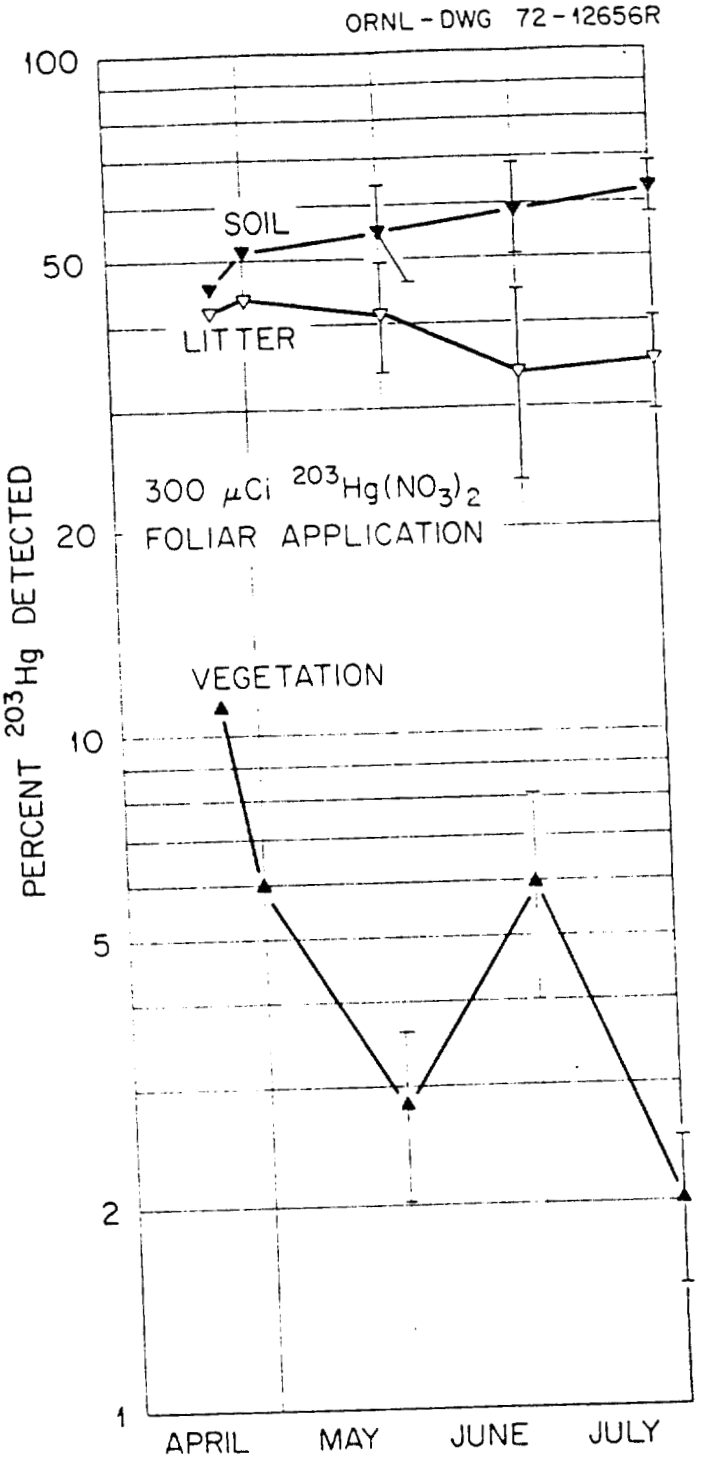

Fig. 4.3.4. Mean percent distribution of ^{203}Hg after tagging by simulated rainfall on fifteen 1-m^2 plots at ORNL, Tennessee, 1972.

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or domestic animals that may feed in such ecosystems.

4.3.3.4 Uptake of Mercury by Bean (Phaseolus vulgaris) Plants

The uptake of mercury by food plants via irrigation represents one pathway in which man may be exposed to this toxic element. To determine the relative mobility of organic and inorganic mercury from water to soil to plants, snap bean seeds were planted in two identical plots which were then watered with solutions of $^{203}Hg(NO_3)_2$ and $CH_3{}^{203}HgCl$, respectively. Mercury solutions were applied four days after planting and bean plants were harvested for ^{203}Hg determinations for a period of eight weeks. Watering solutions contained 97.2 μCi of ^{203}Hg (0.018 ppm Hg) as mercuric nitrate and 102.7 μCi of ^{203}Hg (0.026 ppm Hg) as ,ethyl-mercury chloride, each in one liter of water.

Table 4.3.4 gives the distribution of mercury in plant parts through time. Uptake of methylmercury was consistently higher than inorganic Hg in all plant parts. The most significant result was that ^{203}Hg from the organic form concentrated in the bean fruit by an order of magnitude greater than that from the inorganic form. Mercury in both forms reached concentration levels in beans which were higher than

Table 4.3.4. Mercury uptake by bean plants from soil in East Tennessee, summer 1972

Means, nCi/g dry weight

Sample	Age (weeks)	$CH_3{}^{203}HgCl$	$^{203}Hg(NO_3)_2$
Foliage	2	3.4	0.14
	3	2.2	0.29
	5	2.4	0.41
	8	0.3	0.02
Stems	2	2.1	0.05
	3	1.1	1.1
	5	1.3	0.45
	8	0.1	0.02
Roots	2	4.0	2.4
	3	1.8	1.4
	5	2.4	1.2
	8	1.2	0.01
Pods	8	0.5	0.08
Bean	8	3.1	0.3

those in the solutions applied to the soil. In the bean fruit at

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was the sulfide minerals of Cu and Fe, with some Zn and traces of Pb, Co, and As. No cinnabar (HgS) was detected.[13] The study streams are located in remote sections of the park and are accessible only by foot or on horseback. Historically the area was heavily logged, but all commercial activity was phased out with the establishment of the Park in 1926.

Rainbow trout (Salmo gairdneri) and stonerollers (Campostoma anomalum) were collected at 550 m elevation in all three streams. Rainbow trout were collected at 580 m in Hazel Creek and at 850 and 1225 m in Forney Creek. Eastern brook trout (Salvelinus fontinalis) were collected at elevations between 1150 and 1225 m in all three streams. Rosyside dace (Clinostomus funduloides) and banded sculpin (Cottus carolinae) were collected at 550 m in Hazel Creek. The fish sampled represent two trophic levels, trout, dace, and sculpins being predators and stonerollers feeding on periphyton. The streams are not presently stocked with hatchery fish; however, rainbow trout were introduced approximately 50 years ago. Sediment samples were also collected at 550 m in each of the three streams for mercury analysis.

All fish samples were immediately frozen in Dewar flasks containing Dry Ice (CO_2) for transport to the laboratory. (Since the analytical technique for measuring methylmercury was not in operation at the time, the samples were analyzed for total mercury only. The GCPES technique is now available (section 4.9) and will be used in future studies.) The gastrointestinal tracts were removed from all the stonerollers to avoid the possibility of prejudicing the analyses with mercury-containing sediment or food organisms. Comparative analyses of trout samples were made with the gastrointestinal tract removed and with it present. A comparison was also made of mercury content in trout by dissolving separately for analysis the whole fish and a strip of axial muscle tissue. Under conditions of chronic exposure to mercury, the muscle tissue of fish contains the greatest percent of the total mercury body burden.

Most reports of mercury analyses in fish and other organisms [illegible] wide variability of mercury concentration in tissue.[2,14,15] The actual amounts--nanograms and less--are very small, so a large per

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be a species difference rather than geographical difference (Table 4.3.5). Mercury concentrations have previously been measured in brook and rainbow trout. Brook trout in Canada were found to have a range from 0.08 to 0.13 ppm (wet weight) methylmercury.[15] In Idaho, 16 wild rainbow trout had a mean of 0.31 ppm (wet weight) mercury with a range of < 0.008 to 0.6 ppm, and hatchery reared rainbow trout had a mean mercury concentration of 0.10 ppm with a range of 0.05 to 0.17 ppm.[17]

Earlier studies at ORNL of mercury transport in streams had disclosed mercury concentrations in fish much higher than the putative background levels in the Great Smoky Mountains National Park fish. The ORNL watershed where the stream samples were collected is free from industrial or agricultural discharges (other than fly ash) and the small streams are oligotrophic. However, 38 fish (Rhinichthys cataractae, the long-nose dace) were found to contain a mean concentration of 0.281 ppm mercury. Thirty fish of the same species from a stream in Cades Cove at the 550-m elevation were found to contain a mean concentration of 0.058 ppm mercury (five composite samples of 2 to 11 fish each). The significantly higher ($P \leq 0.01$) concentrations in the ORNL fish indicates that flue gas and/or fly ash may have contributed significantly to these mercury concentrations.

Mercury Stream Tag Experiments. The results of streams tagged with radioactive methylmercury and mercuric nitrate were reported previously.[1] A brief summary of the experiments and additional analyses of the data follows. A small stream within the AEC-ORNL Reservation was tagged with radioactive methylmercury chloride and a similar stream tagged with radioactive mercuric nitrate in an effort to clarify the fate of mercury compounds within natural stream ecosystems. Stream tag experiments, in which a radioactive isotope is added to the water, and the distribution of the activity followed by collecting samples and determining the radioactivity, can rapidly yield quantitative data on transfer rates and bioaccumulators of the isotope. Obviously, these parameters will change from stream to stream, but the qualitative generalizations should hold.

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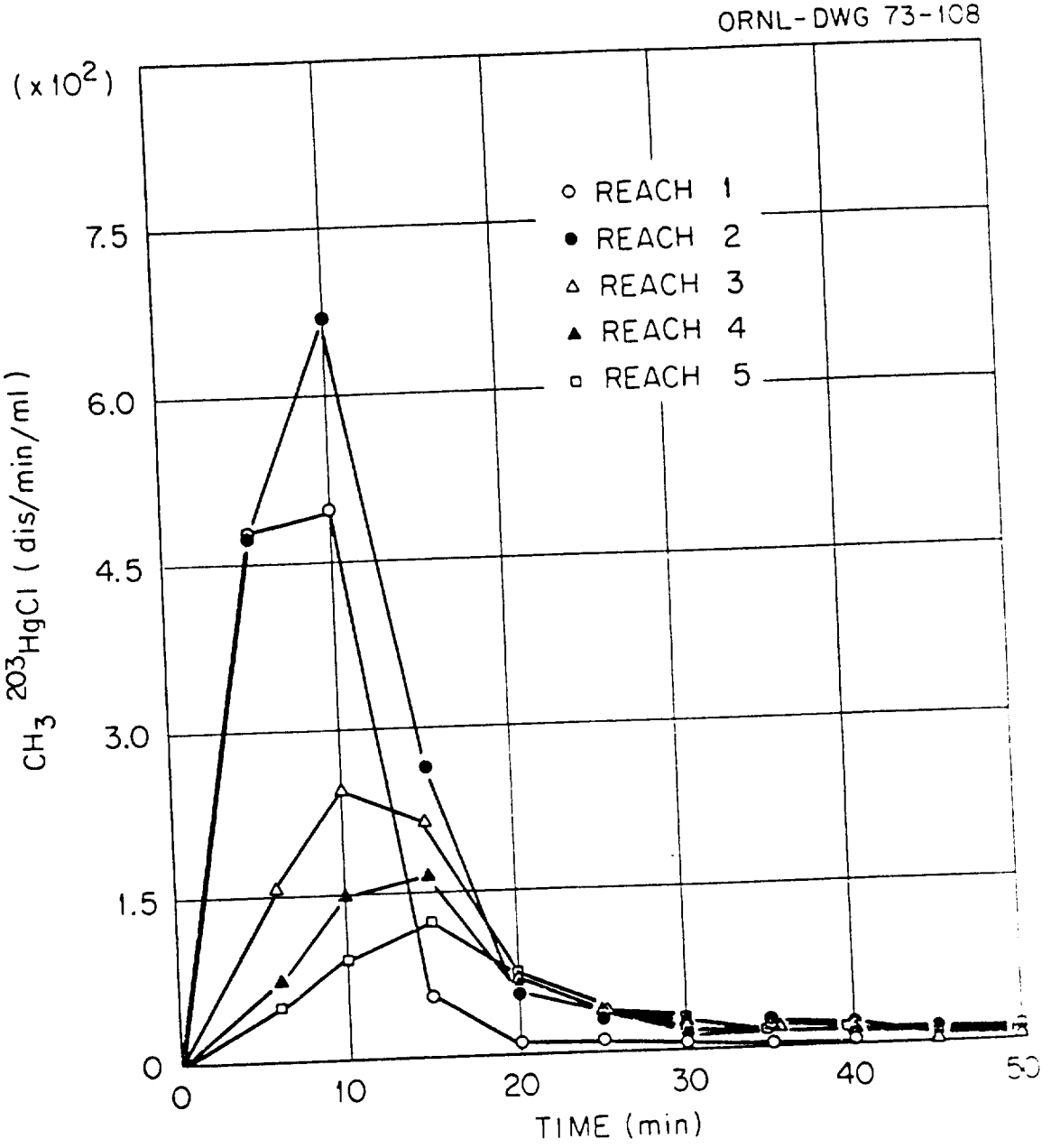

Fig. 4.3.6. ^{203}Hg activity in 5-ml aliquots of stream water at successive downstream sampling stations during CH_3 ^{203}HgCl tagging experiment.

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sediments) on which they grazed, peaked at five days. In the $CH_3{}^{203}HgCl$ experiment, the periphyton peaked at four days, and the snails at 28 days. These data show that inorganic mercury is cycled through the periphyton compartment about twice as fast as methylmercury, while the snail compartment cycles inorganic mercury about five times faster than methylmercury.

Table 4.3.6. Transport of $^{197}Hg(NO_3)_2$ and $CH_3{}^{203}HgCl$ in a stream at ORNL, Tennessee, following spike input

	Station	Day after spike when maximum concentration occurred	
		$^{197}Hg(NO_3)_2$	$CH_3{}^{203}HgCl$
Sediments	1	9	1
	2	7	8
	3	9	8
	4	3	42
	5	2	16
Periphyton	1	1	1
	2	2	1
	3	2	2
	4	2	2
	5	2	4
Snails	1	1	4
	2	1	8
	3	2	12
	4	3	28
	5	5	28

In these oligotrophic waters, Hg^{2+} from methylmercury was concentrated and retained to a greater extent than mercuric ions from $Hg(NO_3)_2$ by all the animals sampled. It was not determined whether mercuric ions from $Hg(NO_3)_2$ were converted to methylmercury. Figures 4.3.8 and 4.3.9 show the concentrations (dpm/g) of radiomercury in the samples of selected stations (all biota present) in each experiment.

Fish acquire and retain methylmercury to a greater extent than other biota and inorganic sediments. During the course of the sample collecting, a few crayfish and salamanders were caught. The concentrations of $^{203}Hg^{2+}$ activity from methylmercury in these species were

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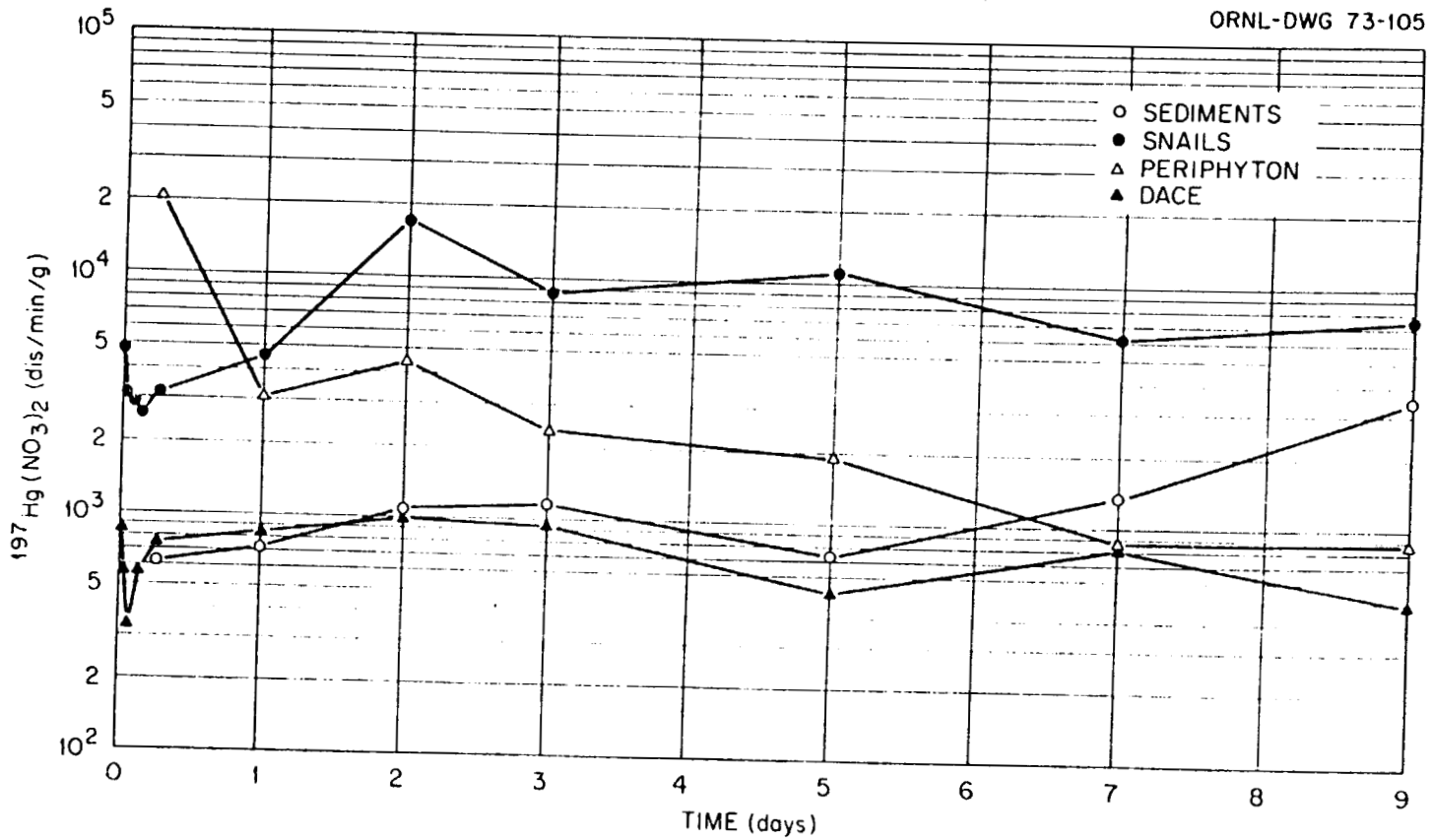

Fig. 4.3.9. Mean ^{197}Hg activity (dis min^{-1} g^{-1}) in stream ecosystem compartments at a selected sampling station (reach 3) during $^{197}Hg(NO_3)_2$ stream tagging experiment. Snails, composite samples, dace, four to six observations; sediments and periphyton, three observations.

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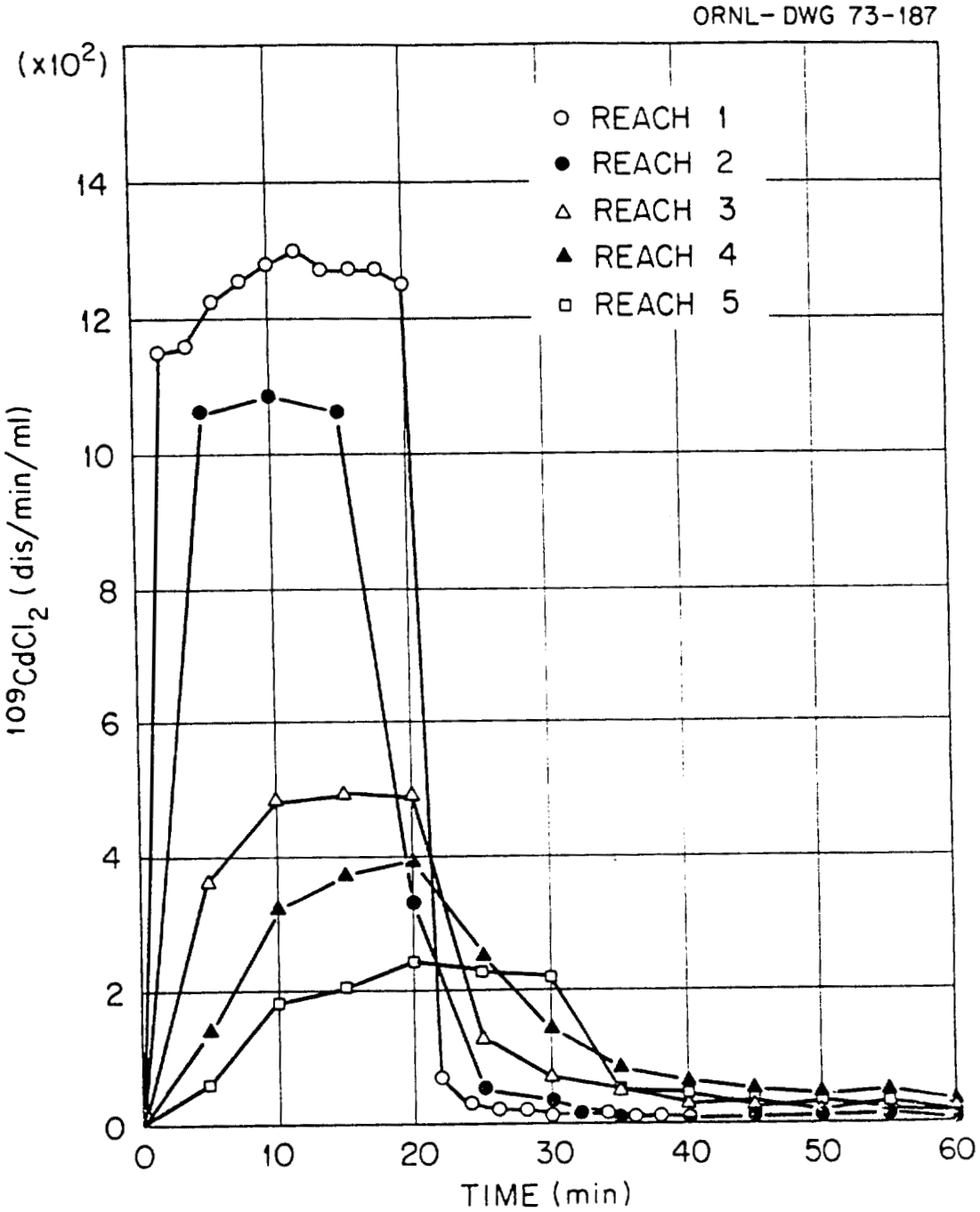

Fig. 4.3.10. ^{109}Cd activity in 5-ml aliquots of stream water at successive downstream sampling stations during $^{109}CdCl_2$ tagging experiment.

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Mercury in Aquatic Food Chains: The accumulation of mercury in fish is one of the most important aspects of the mercury pollution problem, both in terms of ecological effects and human consumption.[2,19,20] Fish acquire body burdens of mercury directly from the water by absorption through the gills and through the food chain,[21] but it is not known if the retention time of mercury differs according to the route of uptake. There have been many analyses of mercury in aquatic organisms, but the trophic dynamics of mercury in aquatic ecosystems remain obscure. Understanding the problem requires consideration of the behavior of both inorganic mercurials and methylmercury. It has been reported that 60-100% of the total mercury in fish tissue is the methyl form, even though the major industrial input to the environment is inorganic or metallic mercury.[2,19,20,21] The conversion of other mercurials to the methyl form by methanogenic bacteria in stream and lake sediments has been reported as the process by which fish accumulate methylmercury. Recently, this explanation as the source of methylmercury in fish has been questioned because of low methylation rates and the failure to detect methylmercury in sediments. Therefore, data on the uptake and retention of methyl and inorganic mercury in fish and data on food chain studies have become increasingly important.

A comparison has been made of the accumulation and retention of CH_3HgCl and $Hg(NO_3)_2$ acquired directly from the ambient water and from the food in the trophic chain: detritus → *Chironomus* larvae → mosquito fish (*Gambusia Affinis*) → largemouth bass (*Micropterus salmodies*) at 25°C. Some of the data which are summarized in Table 4.3.7 have been reported previously. These data were obtained in experiments using radiotracer techniques and stable chemical analyses.

The mechanism of uptake and retention of inorganic mercurials by fish differs from that of organic mercurials.[21,22] Organic mercurials are metabolized in the liver and the Hg^{2+} ions excreted via the kidneys. The cleavage rate of the carbon-mercury bond is slow; consequently the organic moieties are retained longer than inorganic ions, resulting in deposition of organo-mercurials in the nervous system and in muscles.[21] Since fish muscle protein has a higher percent of sulfur-containing amino acids and nitrogen, both of which

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the percentage had decreased to less than 2%. Direct uptake and food chain studies are continuing in order to determine the flow of cadmium through aquatic systems.

Model of Methylmercury Behavior in a Pond Ecosystem: A linear, eight compartment model has been developed to describe the dynamic redistribution of methylmercury in a pond ecosystem (Fig. 4.3.12). The eight compartments were water, sediment, phytoplankton, zooplankton, mosquitofish, bluegill, bass and carp. It was assumed that the methylmercury transfers resulting from predation are donor dependent; that is, the transfer rate is linearly proportional to the amount of methylmercury in the prey compartment. It is also assumed that each of the trophic levels accumulated methylmercury directly from the water, and that mercury loss to the atmosphere was negligible. Eight millicuries of methylmercury was introduced into a 1/4-acre pond to validate the model. The pond was stocked with mosquitofish Gambusia affinis, bluegill Lepomis macrochirus, largemouth bass, Micropterus salmodies, and carp Cyprinus carpio. A dense bloom of zooplankton, mostly Scapholebris kingi, was present during most of the study. The phytoplankton, zooplankton, fish, sediments, and water were sampled periodically and assayed for ^{203}Hg activity. The model gave good qualitative agreement with the general redistribution trends shown by the experimental data. Preliminary analysis indicates direct uptake by trophic levels of methylmercury from water is proportional to size of organism. Preliminary analyses indicate the need to model biomass as well as methylmercury dynamics in cases where there are large fluctuations in trophic level biomass, since the predation rate between trophic levels is a function of the relative sizes of the levels. The predation rate between levels determines the methylmercury transfer rate between levels. More detailed analyses of both the data and the model are currently in progress.

The experimental data, not yet complete, differ quantitatively from the calculated values (Fig. 4.3.13), but the qualitative fit is good. The discrepancies are probably due to some imprecision in measuring the transfer rate coefficients. As more data are

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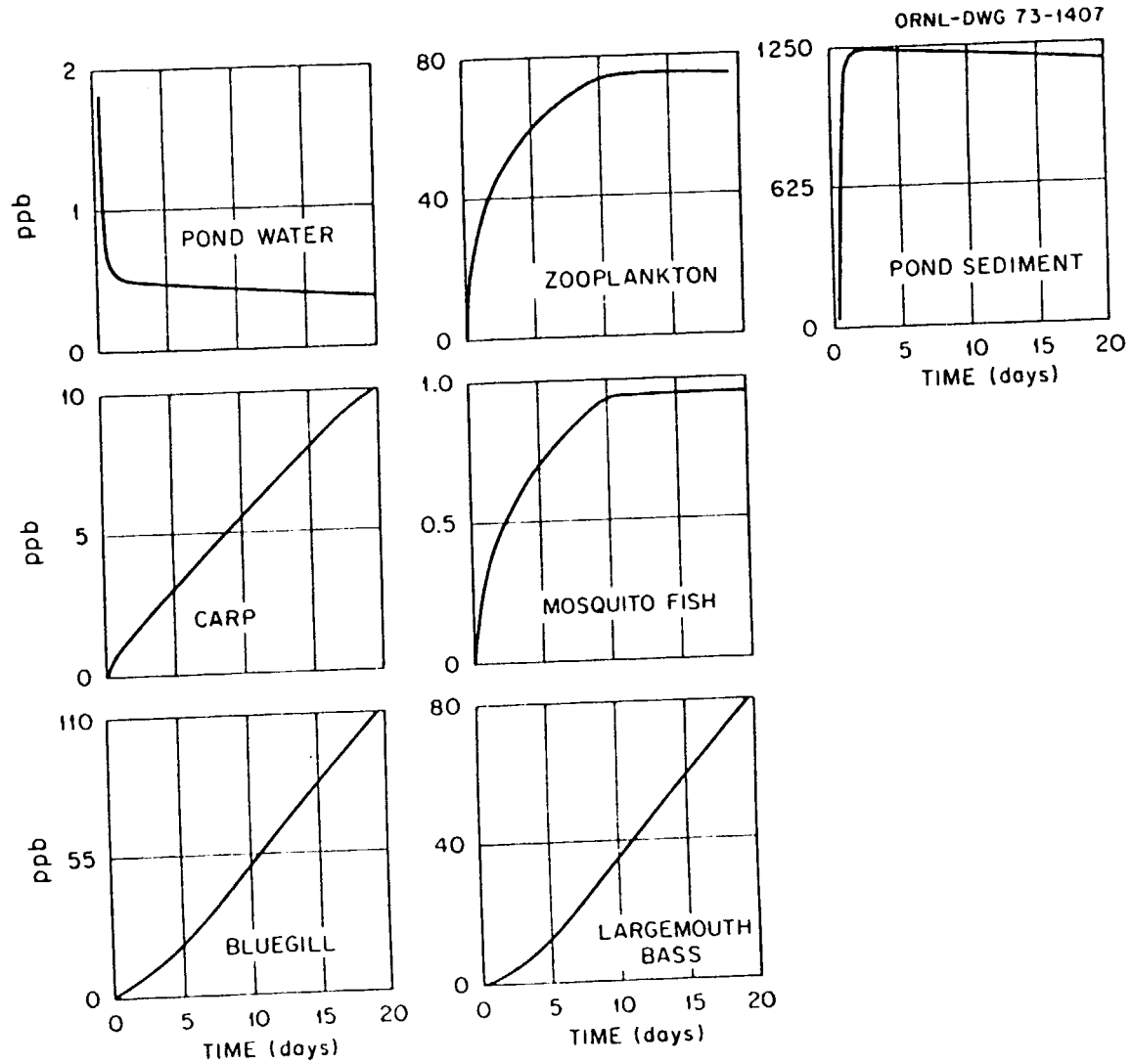

Fig. 4.3.13. Computer simulation curves of the dynamic redistribution of methylmercury added to a pond ecosystem at ORNL.

4.3.7 Potential Application of Work

Ecological data on the movement and concentration of toxic metals in different ecosystems are necessary in order to predict the fate of these substances in the environment. Knowledge of food chain pathways, transfer coefficients and sinks are necessary in taking realistic steps towards pollution abatement. Ecological problems involving toxic materials are current and realistic problems to which students enthusiastically relate. Undergraduates in the NSF summer research program, ORAU summer trainee program, and ORAU undergraduate research program have participated in research on the ecology of toxic metals. A graduate student from the Ecology Institute, University of Tennessee, is completing work on a master's thesis. An ORAU predoctoral fellow from Northwestern University is currently collecting data to determine the influence of temperature and methylmercury concentrations on the kinetics of methylmercury uptake in fish. Two graduate students from the MIT Practice School of Chemical Engineering studied the distribution of methylmercury in an experimental pond.

The background concentrations of mercury and cadmium in ecological samples from the Great Smoky Mountains National Park are being relied upon by the National Park Service. The selection of Cades Cove as the site in Great Smoky Mountain National Park (GSMNP) for heavy metal analysis of vegetation was suggested by Paul F. McCrary, Chief Park Naturalist. The Cades Cove Loop Road is so heavily utilized that the Park Service is concerned about the environmental impact of vehicular traffic. Our preliminary results indicate that vegetation within 1 meter of the pavement is enriched in Pb, Cd, and Zn, but not Hg. These results along with other factors are being considered by the Park Service in their pending decision whether or not to curtail or eliminate automobiles from the loop road.

A GSMNP construction project, the Little River Road, was discovered to be progressing contrary to the environmental impact statement required by NEPA. Soil from a mineralized area had been used for fill material along the stream. Gilbert Calhoun, Assistant Superintendent, GSMNP, requested our advice on possible toxic

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4.3.8 References

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