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Occurrence of Methylated Mercury in a Terrestrial Food Chain[1,2]

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The presence and transfer of methylated mercury (MeHg) in a terrestrial food chain were determined by radiotracer laboratory experiments and chemical analyses of field-collected organisms. [^{203}Hg]MeHg-tagged fescue grass (*Festuca* sp.) at 0.65 μg MeHg/g grass (wet wt) was fed to cotton rats (*Sigmodon hispidus*). The rats had an assimilation efficiency (A_e) for MeHg of 99% and a biological half-life (t_b) of 9.5 days. Control *S. hispidus* and *Festuca* collected from the same populations as the experimental organisms were analyzed for MeHg and found to have, for the *S. hispidus*, 1.4 ± 0.2 ng/g, 1 SE, and 1.3 ± 0.3 ng/g, 1 SE, in skeletal muscle and liver, respectively; and, for *Festuca* (composite sample) 0.6 ng/g. Total mercury (ΣHg) for the same tissues was 119.4 ± 39 ng/g, 1 SE, 34.6 ± 8.9 ng/g, 1 SE; and 71 ng/g (composite sample), respectively. Calculations with the experimental and analytical MeHg values indicated that the feral *S. hispidus* would, at equilibrium, have a whole-body MeHg concentration of 1.89 ng/g. These results, compared with literature reports, indicate that feral rodents have concentration ranges of ~50 to ~160 ng/g for ΣHg, and ~1 to ~7 ng/g for MeHg. Environmental levels of mercury are likely to increase significantly; further environmental measurements and especially appropriate toxicity tests are warranted to assess the importance of MeHg in terrestrial ecosystems.

INTRODUCTION

Information on the distribution and behavior of methylated mercury (MeHg) in terrestrial food chains has been almost entirely limited to episodes of the misuse of MeHg-containing fungicides. Instances of such misuse came rather suddenly to light in the late 1960s and early 1970s when it was realized that granivorous birds in the United States, Canada, and Scandinavia were consuming MeHg-treated seed and were being thereby contaminated to the extent that many of them were unfit for human consumption (Wallace *et al.*, 1971; Benson *et al.*, 1971; Fimreite *et al.*, 1970; Borg *et al.*, 1969; Berg *et al.*, 1966). Other species, especially raptors, were also found to be heavily contaminated with MeHg, indicating MeHg transmission in food chains (Borg *et al.*, 1969; Fimreite *et al.*, 1970). These MeHg fungicides were the same compounds that were responsible for the tragic human poisoning

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distilled water for 10 min, and air-dried for half an hour. Each bundle was radioassayed for ^{203}Hg activity in a 6 × 6-cm through-hole NaI detector coupled to a multichannel pulse–height analyzer. All ^{203}Hg γ-ray counts were compared with a standard prepared from the stock [^{203}Hg]MeHg solution for determination of counting efficiency and normalization of radioactive decay.

The radiotracer-tagged fescue grass was to be exposed to MeHg in such a way as to accumulate levels near to those found by the actual transfer pathways. However, preliminary experiments showed that plants tagged with less than a few hundred nanograms per gram of MeHg (as [^{203}Hg]MeHg) did not result in sufficient ^{203}Hg in the rats for reliable live-count γ-ray measurements. Therefore, we tagged the fescue to levels of ~790 ng/g in order to provide sufficient radioactivity to adequately follow the path of the ^{203}Hg in the rats. This dosage (6.0 ng MeHg/g body wt) is well below the calculated minimum toxic levels using data reported by Spyker *et al.* (1972) for *Mus musculus* (~485 ng MeHg/g body wt, assuming 16.5 g average body wt).

The bundles of tagged fescue were presented to the rats (one per rat). After about 5 hr each rat and the fragments of uneaten food were counted for ^{203}Hg activity. The rats were whole-body counted in an Armac liquid scintillation detector.

A preliminary experiment to determine percentage MeHg in feces and urine was run in which blotter paper was placed below the floor grill in each rat's cage. Fecal pellets and urine soaked blotter paper were counted for ^{203}Hg after 24 hr postfeeding.

Uneaten grass was collected from each cage and radioassayed; activity remaining was subtracted from that originally presented to the rat to get a value for total activity actually ingested. Feces were collected daily from each rat and assayed for activity. In this way, whole-body loss, cumulative loss in feces, and initial activity ingested were recorded, and mass balance of ^{203}Hg could be followed.

Rats and feces were radioassayed daily for 22.5 days postfeeding. The data were subjected to least-squares regression analysis and the biological half-life (t_b) and assimilation efficiency (A_e) were calculated. The rats were sacrificed and dissected on Day 23 post-tag. Liver and skeletal muscles were assayed for radioactivity.

Subsamples of the meadow grass and 16 feral *S. hispidus* were collected in the same meadows as the dams of the experimental animals. Samples of the grass and skeletal muscle and liver were analyzed for ΣHg and MeHg content by cold vapor atomic absorption spectrophotometry (ΣHg) and microwave emission gas chromatography (MeHg). Appropriate analytical (laboratory prepared) and procedural (National Bureau of Standards) standards were also analyzed to insure quality control. No procedural standard currently exists for MeHg. Analytical methodologies employed for these measurements have been reported before (Huckabee *et al.*, 1974; Talmi, 1975).

A MeHg whole-body burden at equilibrium in *S. hispidus* was calculated with the experimental data. The calculations were verified by comparison with the MeHg chemical analysis results.

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TABLE 1

CONCENTRATION (ng/g wet wt) OF ∑Hg, MeHg, AND PERCENTAGE OF ∑Hg AS MeHg IN MUSCLE AND LIVER TISSUE OF *Sigmodon hispidis* AND FESCUE (*Festuca* SP.) CO-OCCURRING NEAR ORNL, TN, AUGUST 1978, AND IN GREAT BRITAIN, 1977

S. hispidus sample	∑Hg		MeHg		Percentage MeHg	
	Muscle	Liver	Muscle	Liver	Muscle	Liver
1	200	32	0.7	0.4	0.35	1.3
2	90	157	1.8	0.7	2.0	0.45
3	144	15	1.0	0.9	0.69	6.0
4	37	15	2.2	3.2	6.0	21.3
5	54	16	1.8	2.1	3.3	13.1
6	32	30	2.6	2.7	8.1	9.0
7	97	41	2.1	1.7	2.1	6.6
8	145	12	0.7	0.9	0.48	7.5
9	51	22	0.4	0.3	0.78	1.4
10	680	15	0.6	0.4	0.09	2.7
11	34	12	1.1	<0.5	3.2	4.2
12	115	11	0.2	0.1	0.17	—
13	34	72	1.6	1.4	4.7	19.4
14	88	108	1.4	0.4	1.6	0.4
15	50	26	3.0	1.8	6.0	6.9
16	60	—	1.1	0.9	1.8	—
$\bar{X}$ ± 1 SE	119.4 ± 39.3	34.6 ± 8.9	1.4 ± 0.2	1.3 ± 0.3	2.6 ± 0.6	7.2 ± 1.9
Clethrionomys glareolus[a]	60 ± 10	60 ± 20	2.9	6.9	4.8	11.5
Apodemus sylvaticus[a]	70(NS)	40 ± 10	2.9	6.9	4.1	17.3
Fescue sp.	71		0.6		0.85	
Fescue sp.[a]	26		—		—	

[a] Bull *et al.* (1977).

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If the ingestion rate of fescue by S. *hispidus* = 30 g/day (Kay and Dunaway, 1962; Kaufman, 1976; Gentry *et al.*, 1975), the MeHg concentration in fescue = 0.6 ng/g, A_e MeHg = 99%, t_h MeHg = 9.5 days, and $\bar{X}$ body wt = 129.2 g, then

$$N_{\text{equilibrium}} = \frac{(30\ \text{g/day} \times 0.6\ \text{ng/g} \times 0.99 \times 9.5\ \text{day})}{0.693 \times 129.2\ \text{g}} = 1.89\ \text{ng/g}.$$

This value—1.89 ng/g—for whole-body concentration of MeHg in *S. hispidus* at equilibrium compares favorably with the measured concentration in muscle (1.4 ng/g) and liver (1.3 ng/g) in the feral *S. hispidus*. The MeHg tissue concentrations at which toxic effects begin to occur in *S. hispidus* are not known.

These results show that MeHg is probably widespread in the terrestrial environment, although at low levels. Since the likelihood of these MeHg levels rising is significant, further measurements of MeHg and especially toxicity tests of MeHg to terrestrial food chain animals are warranted. Toxicity tests, compared with the ecological parameters discussed here, will yield predictive capabilities for the effects of MeHg on terrestrial organisms.

Industries that burn coal and other Hg emitters may be required to monitor Hg in terrestrial ecosystems under the Clean Air Act (42 USC 1857 *et seq.*). Our results indicate that the most efficient and cost-effective regime of monitoring for compliance with Hg standards would be to sample and analyze only primary producers (vegetation), if transfer coefficients in the relevant food chains are known.

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