Contamination of Brown Trout *(Salmo trutta)* by Tributyltin from Timber Treatment Plants

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Total tin and tributyltin (TBT) concentrations were measured in the brown trout, *Salmo trutta*, in 1988 from three river systems that had been contaminated by losses of TBT from timber preservation plants. Mean total tin concentrations in trout muscle ranged from 0.07 to 0.48 mg/kg. In the more contaminated areas approximately 30% of this tin was in the form of TBT. © Crown copyright 1997

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INTRODUCTION

The primary concern arising from the release of tributyltin (TBT) compounds to the environment has been related to the effects of these substances on marine organisms, particularly molluscs.[1] As a result of investigations of these effects, the use of TBT in antifouling preparations is now controlled in many countries. Even so, impacts on gastropods are sitll considered serious in some areas,[2,3] and effects on additional species continue to be reported (e.g. *Littorina littorea*[4]).

Freshwater environments have also been at risk from exposure to TBT through a variety of uses, including antifouling paints,[5,6] industrial discharges, municipal waste waters,[7] domestic and commercial timber treatment and so on.[8–10] Water bodies with long flushing times must be considered at greater potential risk.[17] Effects on freshwater organisms have been more difficult to demonstrate in the field, and greater emphasis has been laid on the chemical detection of organotin residues in freshwater sediment, water and biota (e.g. Ref. 6).

In 1987, the UK legislation prohibited the use of TBT in antifouling products on vessels less than 25 m in length and in aquaculture. The restrictions on the use of TBT were increased in 1990, when the Department of the Environment announced that the use of TBT wood preservatives and surface biocides was limited to industrial processes and professionally applied paste formulations. Despite care being taken in the commercial use of TBT paints, contamination of some freshwater systems sporadically occurred before restrictions were made on use of TBT. This report presents data on the concentrations of tin and TBT in the brown trout (*Salmo trutta*) from three water courses in the Grampian Region of Scotland which have a history of chronic or acute TBT contamination by wood preservatives from timber treatment plants.[12] Brown trout provide important recreational fisheries in Scotland and are present in all three rivers.

METHODS

Sampling locations

The three water courses selected for sampling in October 1988 are described below.

Area A: Tycock Burn/River Lossie system, Elgin, Grampian (Fig. 1)
The Tycock burn is a small stream (catchment area 3.1 km^2) which drains into the River Lossie (catchment area above Kirkhill 266 km^2). At least two major spillages of preservative from a sawmill are known to have occurred in 1983, which both contaminated the ground adjacent to the water course and entered the stream causing acute toxicity to aquatic organisms.[12] Subsequent

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ground contamination through minor spillages occurred over a number of years at the sawmill and another adjacent mill. The formulation of the preservative in use at the sawmill at the time of the survey in 1988 was: pentachlorophenol 1.77%, w/v; tribultyltin 0.44%, w/v; and dieldrin 0.5%, w/v, in a petroleum distillate product. Soon after the survey, the sawmill ceased using TBT-based preservatives (NERPB, personal communication). No fish were found in the Tyoch Burn, but samples were collected at two sites in the River Lossie, Kirkhill (NJ 252626) and Arthur's Bridge (NJ 253673).

Area B: Brodiach/Ord Burn system, Westhill, Aberdeen (Fig. 2)

The Brodiach and Ord burns are both tributaries of the River Dee. Ground contamination by wood preservative used in a timber treatment plant led to chronic low-level pollution of the Elrick Burn and the subsequent water courses joined downstream, namely the Brodiach (catchment area 11.7 km^2), Ord and Leuchar Burns.[12] The active ingredients in the preservative were: tributyltin oxide 1%, w/w; and γ-hexachlorocyclohexane (Lindane) 0.5%, w/w, dissolved in a petroleum distillate product. No fish were found in the Elrick Burn, but were collected at two sites on the Ord Burn: Mill of Brotherfield (NJ 834046) and Ord Dam (NJ 822042).

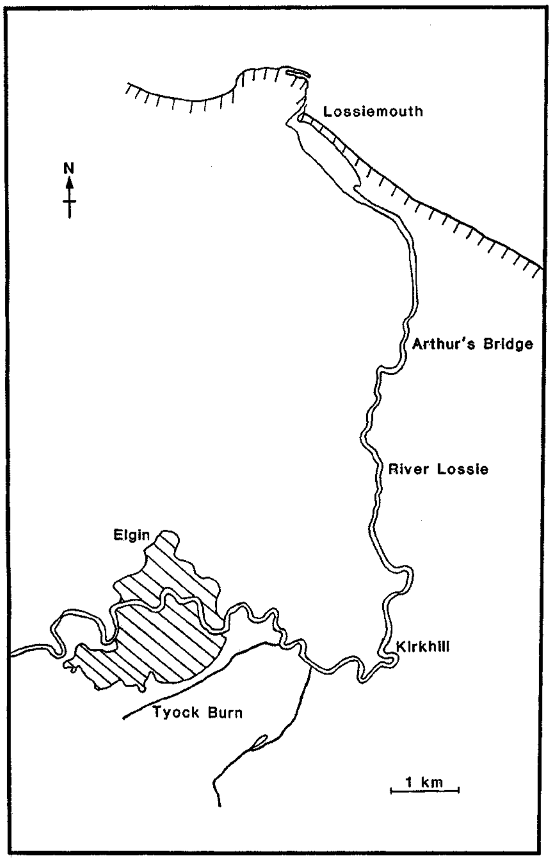

Figure 1 Fish sampling sites in the Tyock Burn area, Grampian.

Area C: Farrochie Burn/River Cowie, Stonehaven, Grampian (Fig. 3)

A major spillage (about 4000 l) of wood preservative occurred in April 1987 from a timber treatment plant on an industrial estate in Stonehaven.[12] The spillage was taken by a surface water drain to a small stream, the Farrochie Burn (catchment area 3.5 km^2), and thence to the River Cowie (catchment area 72.7 km^2) about 0.5 km above its mouth in Stonehaven Bay. A complete kill of fish and invertebrates in the Farrochie Burn, and the short stretch of the Cowie, resulted. Further flushes of the pollutant from the contaminated sewer and the Farrochie Burn were observed after heavy rainfall (NERPB, personal

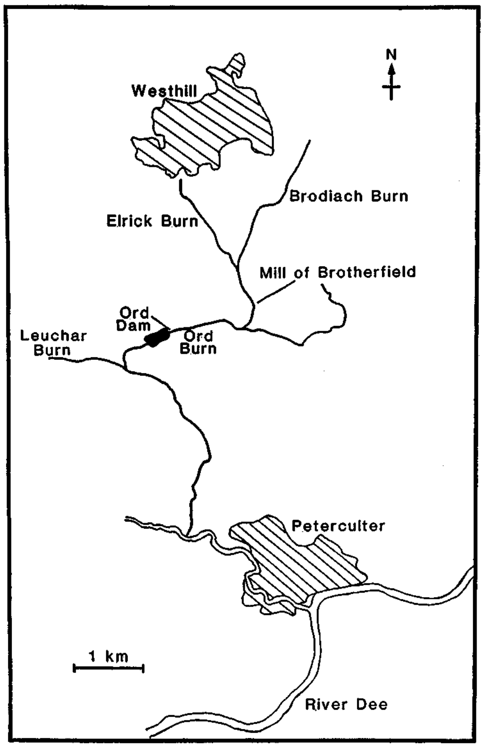

Figure 2 Fish sampling sites in the Brodiach/Ord Burn area, Aberdeen.

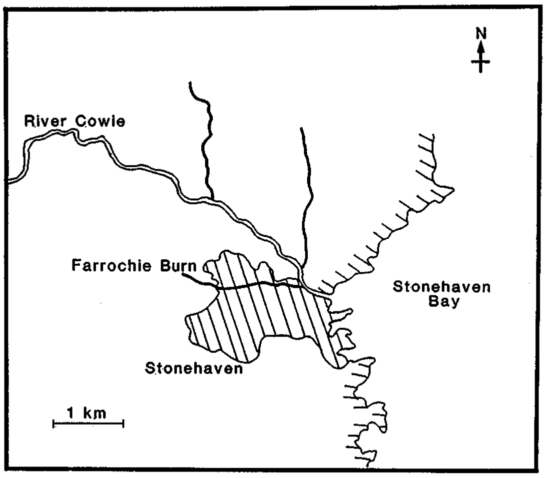

Figure 3 Fish sampling sites in the River Cowie area, Grampian.

communication). The active ingredients in this wood preservative were: tributyltin oxide 1%, w/w; and γ-hexachlorocyclohexane (Lindane) 0.5%, w/w, dissolved in a petroleum distillate product. No fish were found in the Farrochie Burn, but samples were collected in the River Cowie (NO 873865).

Fish capture, preparation and analysis

Lugab backpack electrofishing equipment with an onload of 300 V at the anode, using pulsed direct current at 30 pulses per minute, was used to catch trout at each site. In the laboratory, the length, weight and age of each fish were recorded. Samples of muscle tissue (and liver and kidney from larger individuals) were taken, stored frozen, and subsequently analysed for total tin. Muscle samples from selected trout were analysed for TBT with detection limits of 0.02 mg/kg as total tin or 0.05 mg/kg as TBT by the method of McKie,[13] viz. electrothermal quartz furnace atomic absorption spectrometry after hydride generation.

RESULTS

Almost all the fish caught from all sites were less than three years old, with older fish being found only at Kirkhill (Fig. 1). Weight/length relationships (Fig. 4) indicated a similar growth pattern for fish from all sites.

The mean total tin concentrations (Table 1) in the muscle of trout of all ages from Kirkhill (0.07 mg/kg) and Arthur's Bridge (0.07 mg/kg) in the Lossie system were a factor of 2–5 times lower than that in fish from the other sites. In the Ord Burn, higher tin concentrations were found in fish from the Mill of Brotherfield (mean 0.24 mg/kg) than from the Ord Dam site (mean 0.17 mg/kg). There was a tendency at the more contaminated sites for the highest concentrations to be found in the smaller fish (e.g. Ord Dam, Cowie, Mill of Brotherfield), and the highest mean tin concentration in muscle was found in

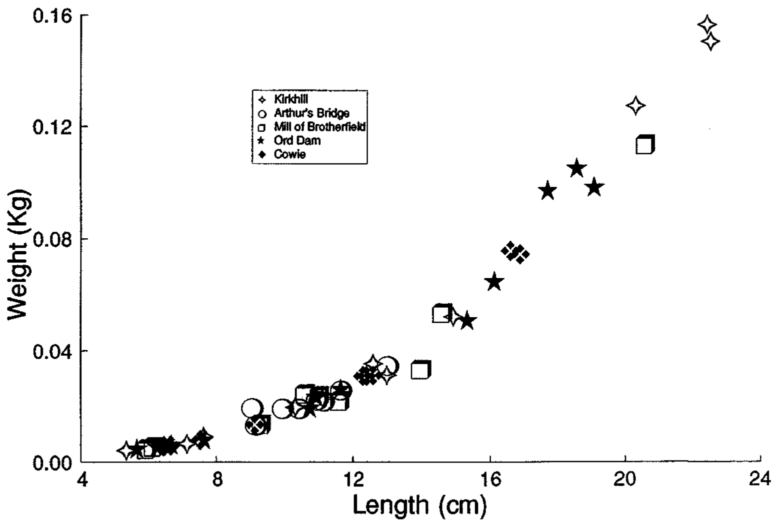

Figure 4 Relationship between the weight (kg) and length (cm) of trout (*Salmo trutta*) from all the survey sites.

Table 1. Trout (*Salmo trutta*) survey; summary of fish data

Location	Age of fish (y)	No. of fish	Mean length (cm)	Mean weight (kg)	Mean total Tin concn (mg/kg wet wt) Flesh[a]	Liver	Kidney
Kirkhill	4+	2	21.25	0.1409	0.05	0.06	0.05
	1+	5	12.28	0.0312	0.08	0.02	—
	0+	4	6.50	0.0054	0.07	—	—
Arthur's Bridge	1+	8	10.53	0.0209	0.07	—	—
Mills of Brotherfield	2+	1	20.50	0.1126	0.28	<0.02	—
	1+	6	12.26	0.0304	0.19	—	—
Ord Dam	2+	4	17.18	0.0791	0.11	0.43	0.19
	1+	7	8.36	0.0127	0.21	0.26	—
Cowie	2+	4	13.45	0.0409	0.21	0.44	0.34
	1+	4	7.30	0.0073	0.48	—	—

[a]Muscle tissue.

one-year-old trout from the River Cowie (0.48 mg/kg).

Total tin concentrations in liver and kidney tissue (Table 1) were higher than in corresponding muscle tissue of the fish from both the Ord Dam and the Cowie sites. Total tin concentrations in the liver, kidney and muscle tissue of fish from the Kirkhill site were similar, suggesting a lower level of TBT contamination in this area.

TBT concentrations in muscle tissue (Fig. 5) tended to reflect the pattern of distribution of total tin concentrations, and were highest in fish from the Cowie (0.20–0.35 mg/kg expressed as TBT) and from the Mill of Brotherfield (0.1–0.35 mg/kg TBT). TBT concentrations in fish from Kirkhill and Arthur's Bridge (River Lossie) were below the detection limit (0.05 mg/kg TBT).

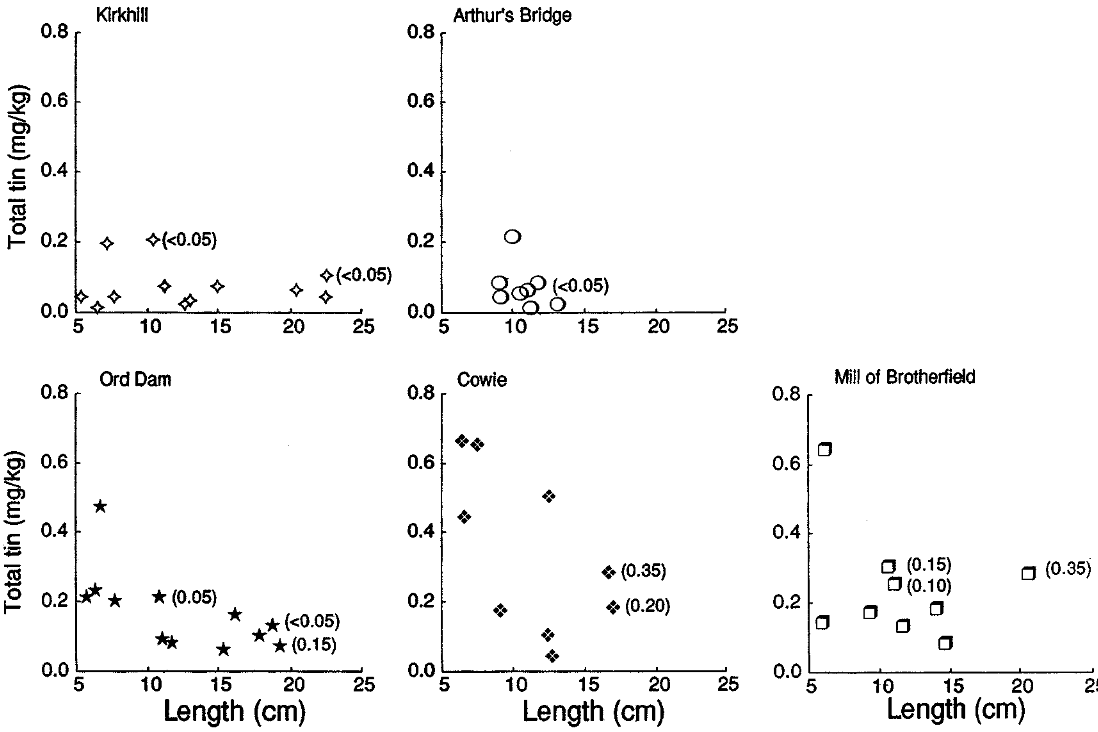

Figure 5 Total tin and TBT concentrations (mg/kg wet weight) in muscle tissue of trout (*Salmo trutta*) from each survey site.

DISCUSSION

Temporal changes in the distribution of trout populations in particular rivers are difficult to predict accurately, since behaviour is dependent upon a large number of variables such as current velocities, stream discharge rate and food abundance. In general, adult fish move in late autumn to locations where there is well-aerated gravel, in order to spawn. Once spawning is complete, the adult fish drop back to their favoured habitats of mid-water pools. The eggs over-winter in the gravel, hatching in the early spring, after which the young fish may spend approximately two years around the redd before moving downstream to find deep water pools where they tend to remain for long periods of time. Some young fish, however, have been known to spend their entire lives in the area where they were born. Owing to the habit of remaining at one location in the river for a long period of time, trout may be a useful tool, both for monitoring levels and for pinpointing areas of contamination.

The patterns of TBT input in survey areas A (River Lossie) and C (Cowie) were similar, in that both areas had been subject to acute and chronic pollution incidents over a long period of time.[12] It might therefore be expected that the tin and TBT levels measured in fish from these areas would be similar. Fish caught from the Cowie, only 1.5 km from the site of TBT spillage, contained the highest muscle concentrations of both total tin (mean 0.35 mg/kg) and TBT (mean 0.28 mg/kg) found in the surveys. Fish caught from the River Lossie, however, contained low concentrations of tin (0.05–0.08 mg/kg), and TBT was not detectable. Since TBT persists in freshwater with a half-life of six weeks to a few months,[14] the low concentrations of tin in fish from the River Lossie suggest that the effluent from the sawmill had been sufficiently diluted before reaching these areas. The nearest sampling site, Kirkhill, was over 4 km from the sawmill, and approximately 2 km downstream from the point where the Tyock Burn joins the River Lossie, at which point great dilution occurs. Although the Farrochie Burn is of a similar size to the Tyock Burn (catchment areas of 3.5 and 3.12 km^2 respectively), the River Cowie is much smaller (catchment area 72.7 km^2) than the Lossie (catchment 266 km^2), and the survey site closer to the TBT source. Less dilution of the contaminant would therefore be expected in the Cowie than in the Lossie.

Examination of the data from the Brodiach/Ord Burn, area B, indicates that both tin and TBT levels were higher in fish from the Mill of Brotherfield (mean concentrations: 0.26 mg/kg total tin and 0.20 mg/kg TBT), which is approximately 2 km from a timber treatment plant, than in fish caught at Ord Dam (mean concentrations: 0.16 mg/kg total tin and 0.07 mg/kg TBT), approximately 3.5 km from the plant. Again, the distance of the survey site from the source of contamination would partly account for the lower tin concentrations found at Ord Dam. Another important factor, however, is that the contaminant released into the Elrick Burn would have been diluted by the Brodiach Burn at the first sampling site, and additionally by the Ord Burn at the second site.

Chliamovitch and Kuhn[45] have demonstrated two toxic mechanisms of TBT in trout, one occurring at high concentrations, which causes rapid destruction of the gill epithelium, and the other at lower concentrations, which causes inhibition of the main metabolic pathways. None of the fish sampled in this survey showed visible signs of severe TBT pollution, such as excessive skin mucous secretion, or damage to the gills or eyes, although fish from all of the sites closest to the sawmills and timber plants had elevated levels of TBT within their muscle tissues. Subsequent work on fish has investigated the impact of TBT on hepatic microsomal monooxygenase systems, and sodium/proton exchange in erythrocytes.[16,17] Histological and histochemical changes in various tissues were observed after exposure of rainbow trout to 0.6–4.0 μg/l TBTO.[18]

Concentrations of TBT in the muscle tissue of the trout from the Cowie (mean 0.28 mg/kg) and the Mill of Brotherfield (mean 0.20 mg/kg) were similar to the lower concentrations found in the muscle tissue of farmed salmon raised in cages treated with TBT-based antifoulants (range 0.2–1.0 mg/kg, cf. Ref. 19). According to Schweinfurch and Gunzel,[20] these concentrations were unlikely to have been of public health concern. However the restrictions on use of TBT in timber treatment plants should have ensured that continuing contamination is unlikely.

The distribution of tin between tissues was consistent with the findings of Martin *et al.*[21] who demonstrated that TBT partitions in trout in three areas, in decreasing concentrations: the peritoneal fat; the liver, kidney and gall bladder; and the other tissue, including muscle. Butyltin compounds are known to accumulate in lipid-rich liver tissue (cf. Refs 19, 27), where detoxification mechanisms operate; and in kidney tissue, prior to excretion.

CONCLUSIONS

Infrequent spillages of preservative containing tributyltin compounds and other pesticides have led to acutely toxic effects on aquatic fauna in adjacent water courses. Lower-level chronic inputs, from contaminated ground or drainage systems, are more common. Mean concentrations of total tin in trout muscle in three contaminated water courses ranged from 0.07 to 0.48 mg/kg. Tribultyltin made up approximately 30% of the total tin in trout from contaminated areas, indicating that degradation of TBT was occurring.

The effects on aquatic fauna resulting from both infrequent spillages and lower-level chronic inputs of preservative containing tribultyltin compounds highlights the part dilution plays in dispersion of wastes. The importance of dilution factors should be emphasized in planning applications and also in environmental impact assessments.

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