



# **Surveys of the River Taff (Cardiff) and associated watercourses**

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**Surveys of the River Taff (Cardiff) and associated watercourses**

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## Executive summary

Isotopically labelled compounds, predominantly tritium ( $^3\text{H}$ ) and carbon-14 ( $^{14}\text{C}$ ), are produced by Amersham plc at their plant in Cardiff, Wales, for use in life science research. Gaseous and liquid wastes are released into the environment under authorisation from the Environment Agency Wales. The liquid effluents are discharged to the Ystradyfodwg and Pontypridd (Y&P) trunk sewer and conveyed into the Severn estuary. The present sewerage system at Cardiff is old and inadequate for dealing with present day volumes of sewage. These factors can result in discharges of crude sewage (hence liquid effluent) into the River Taff, downstream of the confluence with Whitchurch Brook, via the Combined Sewer Overflow (CSO) system. Although a significant body of evidence has now accumulated concerning the impact of the discharges upon the marine environment, data concerning local freshwater bodies are sparse. The objective of the present investigation was to determine  $^3\text{H}$  and  $^{14}\text{C}$  activities in relevant watercourses. Given that levels of  $^3\text{H}$  in the discharges are typically two orders of magnitude greater than  $^{14}\text{C}$ , greater resources were directed towards the former nuclide.

A comprehensive set of indicator samples was collected by independent contractors from the River Taff, the Whitchurch Brook tributary, Glamorgan Canal, River Ely and Cardiff Bay on eight separate occasions, between August 2001 and December 2003. Radiochemical analyses were carried out at the CEFAS Lowestoft Laboratory.

Tritium activities in sediments, invertebrates and fish obtained from 'control' sites (< 10 km to the north and west of the Amersham plc plant) were below the limit of detection. However, the aerial emissions did have a noticeable impact upon two small watercourses close to the Amersham plc plant. Levels in fish from Whitchurch Brook and Glamorgan canal were in the order of  $10^3$  Bq/kg wet weight. In comparison to the small watercourses, the impact of aerial emissions upon levels in the River Taff was minor, presumably due to the considerably greater dilution factor produced by higher water flows.

The impact of liquid waste discharges, via the CSO, upon  $^3\text{H}$  activities in water from the River Taff was barely detectable. In contrast, levels in sediment, invertebrate and fish samples were clearly enhanced. Although considerably less marked, similar trends were apparent in the  $^{14}\text{C}$  data.  $^3\text{H}$  activities in sediment immediately upstream of the CSO were below the detection limit. Downstream, levels were above the detection limit. There was, however, very large scatter in the data limiting the use of sediment as an indicator material in a monitoring programme. There was no overall trend in sediment activities over the study period. Invertebrate samples obtained immediately upstream of the CSO contained measurable amounts of  $^3\text{H}$ , as a result of the aerial emissions. Downstream, levels were enhanced by ~ 10 fold. To a first approximation, activities remained essentially constant throughout the study period. Levels of OBT in invertebrates at all sites were typically between one and two orders magnitude greater than those observed in the sediments.

The results for  $^3\text{H}$  in fish affected by CSO discharges indicated a complex pattern of behaviour. The information obtained from analysis of bulked samples in the first two

surveys (in 2001) was limited, as some species exhibited large variations in levels between individual fish. Analyses of individual fish indicated the variability was greatest for bullheads. Plots of  $^3\text{H}$  concentration versus fish length demonstrated a noticeable discontinuity in levels between the smallest and largest bullheads. An unequivocal interpretation of the data was hampered by lack of specific information concerning increases in fish length concomitant with age in the River Taff. Nevertheless, the most plausible explanation for the observed behaviour is the presence of three separate age groups in the sample, containing different  $^3\text{H}$  activities. The median concentration in (2+) bullheads (spawned in or before 1999) was an order of magnitude greater than that in 0+ bullheads spawned in 2001. The extreme variability, due to the presence of bullheads spawned in or before 1999, was absent in subsequent surveys as they are a short-lived species (typically < 4 years). The impact of liquid wastes entering the River Taff from the CSO appears to have markedly reduced in 2001 compared with that in 1999 and preceding year, although the reason is not known. The impact upon short-lived fish species such as bullheads is now relatively minor. Levels in some longer-lived species, such as eels, remain elevated in older fish, as a legacy of historically greater releases. Due to changes in operational practices (since April 2003), it is reasonable to anticipate future decreases.

## **1. Introduction**

Isotopically labelled compounds are produced by Amersham plc at their plant in Cardiff, Wales for use in life science research. The predominant isotopes are tritium ( $^3\text{H}$ ) and carbon-14 ( $^{14}\text{C}$ ). Gaseous and liquid wastes are released from the plant into the environment under authorisation from the Environment Agency Wales. The liquid effluents are discharged to the Ystradyfodwg and Pontypridd (Y&P) trunk sewer and conveyed to the Severn Estuary. At the beginning of this investigation, the sewage discharged to sea only received preliminary treatment (comminution). Since March 2001, it has been fully treated at the Cardiff East Waste Water Treatment Works and released into the Severn Estuary via a 3.8 km long sea outfall. The sewerage system in Cardiff is old and is inadequate for dealing with present day volumes of sewage. Combined Sewer Overflows (CSOs) exist to act as relief valves for sewage systems, they are designed to discharge during periods of heavy rainfall to avoid damaging the sewer pipes and prevent flooding of properties. On the River Taff, the CSO in Aberporth Road enters a culverted section of Whitchurch Brook (the open sections of the brook in the village) and then flows into the River Taff between Llandaff weir and Blackweir. Thus, although there is no direct discharge of liquid effluent from the site to the River Taff, these factors can result in discharges of crude sewage – and hence liquid after effluent – into the river.

The total quantities of  $^3\text{H}$  and  $^{14}\text{C}$  released from the plant have varied significantly with time. For example, following the commencement of very minor discharges in December 1979, annual releases of liquid  $^3\text{H}$  increased to a peak value of  $\sim 820\text{TBq}$  in 1986/1987. Thereafter, the liquid  $^3\text{H}$  discharges reduced such that in the 1990s

releases were ~ half the maximum value. In response to queries regarding the impact of the  $^3\text{H}$  discharges upon the Severn estuary, in May 1998 the company greatly reduced releases by retention of some of the concentrated tritiated waste (Robinson, 1999). Overall releases of  $^3\text{H}$  were reduced by ~ 4 fold. It is also worth noting that the levels of  $^3\text{H}$  in the discharges have always been significantly greater than  $^{14}\text{C}$ . Successful efforts to further reduce discharges of both radionuclides are ongoing (Table 1).

Table 1

Chronology of total annual  $^3\text{H}$  and  $^{14}\text{C}$  discharges from the Amersham plc plant at Cardiff since 1997. Data taken from RIFE reports.

Year	$^3\text{H}$ discharge (TBq)		$^{14}\text{C}$ discharge (TBq)	
	Liquid effluent	Aerial emissions	Liquid effluent	Aerial emissions
1997	473	640	1.3	3.0
1998	277	560	1.2	2.6
1999	105	500	1.2	2.1
2000	87	503	0.60	1.6
2001	67	554	0.22	1.4
2002	60	401	0.21	1.9
2003	30	589	0.14	1.7

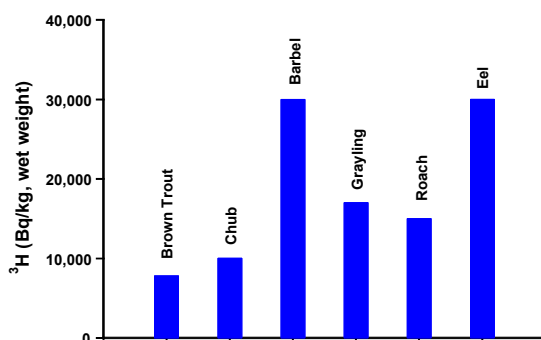
Due to the nature of the work carried out the Amersham plc plant, the composition of the  $^3\text{H}$  and  $^{14}\text{C}$  wastes are complex and contain a wide range of organic compounds. It has been estimated that the tritiated organic compounds historically (pre-May 1998)

formed a minor proportion (< 5 %) of the total liquid  $^3\text{H}$  discharge (Amersham plc, per. comm.). In April 2003, the organic portion of the tritiated discharge was reduced by around 60%.  $^3\text{H}$  in the gaseous (aerial) discharges is operationally classified into “soluble” and “insoluble” forms. The majority (about 80 %) is in the ‘insoluble’ form and consists large of  $^3\text{H}_2$  (gas). The ‘soluble’ form is predominantly  $^3\text{H}_2\text{O}$  vapour.

Although a significant body of evidence has now accumulated concerning the impact of the releases upon the marine environment (Robinson, 1999; Rowe et al., 2001; FSA and SEPA, 1999-2003), prior to the present investigation, only limited data (Fig. 1) were available concerning radionuclides in fish from local freshwater bodies. Results for  $^3\text{H}$  in fish, taken from the RIFE-6 environmental monitoring report (FSA and SEPA, 2001) are provided in Fig. 1.

Figure 1

Tritium activity in fish from the River Taff between Llandaff and Blackweir (August 2000).



The data in Fig. 1 indicate total  $^3\text{H}$  activities in fish ranged from 7800-30,000 Bq/kg wet weight with an average value of  $\sim 18,000$  Bq/kg wet weight. The results clearly

illustrated that levels were significantly elevated above background and of similar magnitude to those observed in fish and shellfish in the Severn estuary. The radiological impact of the elevated levels is small, because  $^3\text{H}$  is a soft  $\beta$ -emitter ( $E_{\beta\text{max}} = 18.6 \text{ keV}$ ) and has a relatively short half-life ( $t^{1/2} = 12.43 \text{ years}$ ); the dose to the most exposed group of fish and shellfish consumers including external radiation was estimated to be 0.064 mSv or 6 % of the principal dose limit for members of the public of 1 mSv (FSA and SEPA, 2001). The primary objective of the present investigation was to assess the relative contribution of the aerial and liquid discharges to  $^3\text{H}$  assimilation in aquatic organisms in selected freshwater bodies, within the vicinity of the Amersham plc plant. In addition, a secondary objective was to assess the impact, if any, of the much smaller  $^{14}\text{C}$  discharges.

## **2. Sampling and Analysis**

### **2.1 Sampling**

A range of indicator samples were collected from the River Taff, the Whitchurch Brook tributary, Glamorgan Canal, River Ely and Cardiff Bay to provide surveillance data (see Fig.2 and Table 2). Samples of riverwater (one litre) and surface sediment were collected from a variety of sites along the River Taff on eight separate occasions, between August 2001 and December 2003. The first four collections were undertaken by the SE Wales Environment Agency appraisal team. Subsequently, this role was carried by Salveson-Fox Hydrosurveys.

There are several physical barriers across the River Taff, including Blackweir, Llandaff Weir, Radyr Weir and Treforest Weir. The three lowermost weirs are all fitted with fish passes which improve fish migration. However, the weir at Treforest serves as a complete barrier to the upstream passage of fish and limits the natural distribution of migratory salmonids. To take account of the physical barriers, individual sections of the river were classified into separate 'reaches' (Table 2).

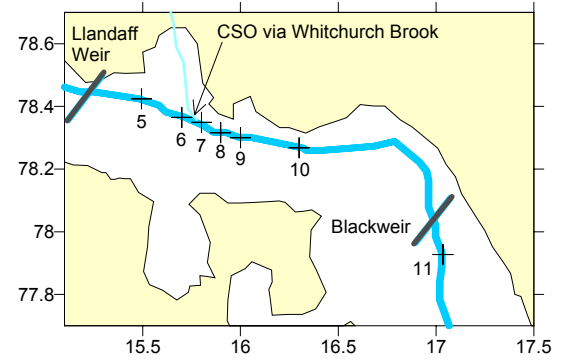
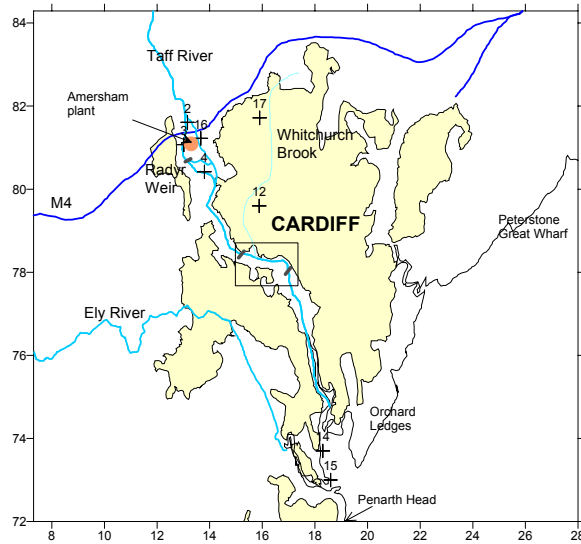
Table 2

Summary of sample sites, radionuclide sources and type of samples collected

Reach	Site Number	Site	Sources	Samples collected
R1	1	upstream of Treforest Weir, acting as a 'control' site	May be subject to aerial emissions	Water, sediment, fish, invertebrates
R2	1a	River Taff at Taffs Well acting as a 'control' site	Aerial emissions only	Water, sediment, invertebrates,
R2	2	upstream of surface drain discharge from Amersham plc	Aerial emissions only	Water, sediment
R2	3	immediately downstream of surface drain discharge	Aerial and site surface water run-off	Water, sediment
R3	4	immediately d/s of Radyr Weir	Aerial and site surface water run-off	Water, sediment, fish, invertebrates,
R4	5	immediately downstream of Llandaff Weir	Aerial and site surface water run-off	Water, sediment
R4	6	immediately upstream of confluence with Whitchurch Brook	Aerial and site surface water run-off	Water, sediment
R4	7	immediately downstream of confluence with Whitchurch Brook	Aerial, site surface water run-off, Aberporth CSO discharge	Water, sediment
R4	8	approx. 100 metres downstream of confluence with Whitchurch Brook	Aerial, site surface water run-off and Aberporth CSO discharge	Water, sediment
R4	9	approx. 200 metres downstream of confluence with Whitchurch Brook	Aerial, site surface water run-off and Aberporth CSO discharge	Water, sediment
R4	10	approx. 300 metres downstream of confluence with Whitchurch Brook	Aerial, site surface water run-off and Aberporth CSO discharge	Water, sediment, invertebrates, fish
R5	11	immediately downstream of Blackweir	Aerial, site surface water run-off, Aberporth CSO discharge	Water, sediment, invertebrates, fish
R6	12	upstream of weir at top end of culverted section of Whitchurch Brook	Aerial emissions only	Water, sediment, invertebrates, fish
R3	16	Glamorgan Canal, adjacent to Amersham plc plant	Aerial emissions only	Water, sediment, fish
R7	13	River Ely upstream of St Fagans, acting as a 'control site'	Aerial emissions only	Water, sediment, invertebrates, fish
R8	14	Cardiff Bay, undredged channel	Aerial, site surface water run-off, Aberporth CSO discharge and historical sediment inputs from the Severn Estuary	Sediment
R8	15	Cardiff Bay, undredged channel	Aerial, site surface water run-off, Aberporth CSO discharge and historical sediment inputs from the Severn Estuary	Sediment

Figure 2

Location of sampling sites along River Taff and elsewhere.



b) Inset. Detailed map to indicate closely spaced sites in River Taff between Llandaff and Blackweir.

a) Map of whole area excluding 'control' sites at Treforrest (site 1) and Ely river (site 13). Rectangle indicates area covered by inset (Fig. 2b).



d) Electrofishing at site 4

Cardiff Bay encompasses the estuaries of two of the main rivers in South Wales – the Taff and the Ely. As of 4<sup>th</sup> November 1999, a barrage has impounded Cardiff Bay by sealing it off from the sea to create a 200 ha lake that is no longer influenced by the tide. The barrage embankment consists of an 800 m long embankment constructed from sand and rock and a 300 m concrete section containing the locks, sluices, fish pass and control building ([www.bensalter.welshnet.co.uk/barragefacts.html](http://www.bensalter.welshnet.co.uk/barragefacts.html)). Five large sluice gates release the flow of the Taff and Ely rivers out of the Bay. The barrage incorporates a specifically designed fish pass to allow salmon and sea trout to return to the rivers to breed.

As available, a range of fish and invertebrates were collected from selected sites in August and September 2001, September 2002, July and December 2003. This was achieved by electrofishing and kick sampling, respectively. Unfortunately it was not possible to obtain samples of all species from each site. Coverage was extended to include vegetation where available in latter surveys. These samples consisted of riverweed from the River Taff and reeds from Glamorgan Canal.

The invertebrates were separated from weed and mud by a combination of sieving and hand picking. Consequently, only small quantities of material (< 10 g) were available for analysis. Individual invertebrate species were combined and preserved in formalin. All samples were delivered in a fresh refrigerated or frozen condition to the CEFAS Lowestoft laboratory for further processing and analysis.

## 2.2 Sample analysis

Water samples were filtered through a tared 0.45µm membrane filter to separate dissolved and particulate phases and to determine the suspended particle concentration. Due to the small weight of particulate material, it was not practical to determine the  $^3\text{H}$  concentration on suspended particles. With respect to fish, the preparation procedures varied between small species (stone loach, bullhead and minnow) and large species (brown trout, roach, eel). Small species were analysed whole, whereas fillet material (combined muscle and skin) was analysed for large species.

Tritium in biological samples is usually categorised as free water tritium ( $^3\text{H}_2\text{O}$ ) and tissue-bound tritium, where the latter means organically bound tritium (OBT) (Bruner, 1973; Hisamatsu et al., 1990). Assay of wet material yielded information concerning the total  $^3\text{H}$  content (i.e.  $^3\text{H}_2\text{O} + \text{OBT}$ ). The OBT content was operationally defined as the  $^3\text{H}$  associated with material from which the water had been physically removed by drying to constant weight at 40°C. Samples were weighed before and after drying to determine the percentage dry solids content (% D. S.). Due to the potential for OBT decomposition during the drying process, and uncertainty of its form, there is clearly some imprecision in the speciation procedure. The values for OBT, therefore, should not be considered totally quantitative, but of use for trend identification.

Tritium assay (wet and dry samples) was achieved by oxidation in chromic acid (a mixture of potassium dichromate, sulphuric acid and phosphoric acid). Using this method (the so-called Walkely-Black method), organic material is oxidised to carbon

dioxide and water (Nelson and Sommers, 1982; Schreier et al., 1999). Water collected as a result of the oxidation process (containing  $^3\text{H}$ ) was distilled into a cold trap. The water was distilled a second time (under alkaline conditions) to ensure chemical separation from all gravimetric and radiometric interference. Subsamples of water distilled from the oxidation process were assayed for  $^3\text{H}$  using a Packard Tri-Carb 2550 TR/LL liquid scintillation counter.

As for  $^3\text{H}$ ,  $^{14}\text{C}$  assay was achieved by oxidation of dry material in chromic acid. Carbon dioxide liberated as a result of the oxidation process (containing  $^{14}\text{C}$ ) was trapped in caustic soda solution and then precipitated as  $\text{CaCO}_3$ . The  $\text{CaCO}_3$  was filtered into a pre-weighed sintered glass crucible washed, dried and reweighed to determine the stable carbon content of each sample. A subsample of the  $\text{CaCO}_3$  was suspended in liquid scintillation cocktail and assayed for  $^{14}\text{C}$  using a LKB Wallac 1217 Rackbeta liquid scintillation counter.

### 2.3 Statistical analysis

Statistical analyses of data were undertaken to assess, amongst other factors, temporal and spatial variation. The data were modelled in terms of one or more median or average values and calculations made of the variance between observations and the appropriate model value(s). Calculations of the F probability distribution (so called p values) were made to assess whether data were significantly better fitted by the simple (e.g. no temporal variation) or more complex models (e.g. including temporal variation). If p values were less than 0.05, the difference between models was statistically significant (at the 5 % level).

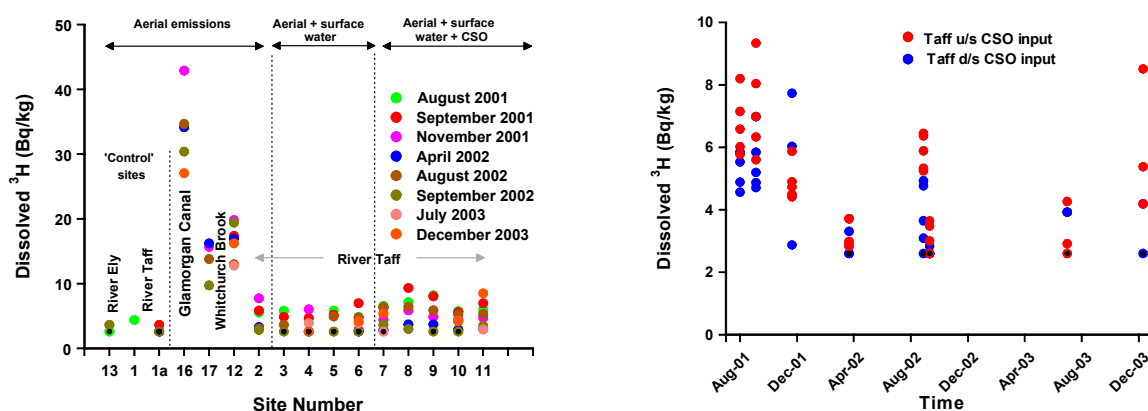
### 3. Results

#### 3.1 Filtered waters

A preliminary investigation indicated  $^{14}\text{C}$  was below the detection limit of the method used in the present study, whereas levels of  $^3\text{H}$  were measurable. Data showing the variability in levels of dissolved  $^3\text{H}$  in the samples of water is given in Figs. 3a and 3b. These values are activities of total dissolved  $^3\text{H}$  (i.e.  $^3\text{H}_2\text{O} + \text{OBT}$ ).

Figure 3

Dissolved tritium in riverwater



a) Spatial variation. Black dots indicate activity  $<2.6$  Bq/kg.

b) Temporal variation in River Taff (sites 2-11)

Levels at the 'control' sites were in the range  $<2.6$ - $4.4$  Bq/kg with an median value of  $2.6$  Bq/kg (Fig. 3a). The greatest activities ( $27$ - $43$  Bq/kg) were observed in samples from Glamorgan Canal (site 16), adjacent to the Amersham plc plant. Levels at sites 12 and 17 in Whitchurch Brook,  $\sim 3$  km distant, from the Amersham plc plant were in

the range 10-20 Bq/kg. The source of the elevated activities in these two water bodies is solely aerial emissions. Levels in the River Taff close to the Amersham plc plant (within 4 km), but upstream of the confluence with Whitchurch Brook (sites 2-6), were slightly elevated above those at 'control' sites (activities were in the range <2.6-7.7 Bq/kg), also as a result of the aerial emissions. The median concentration (4.6 Bq/kg) was ~80 % above that for the 'control' sites. The impact in the River Taff was less than that in the smaller waterbodies, presumably because the dilution factor is greater in the fast flowing river. Downstream of the confluence with Whitchurch Brook (sites 7-11), levels in the River Taff were in the range <2.6-9.3 Bq/kg with an median value of 5.3 Bq/kg. Increases in activities downstream of the CSO overflow were discernable on just four out of the eight surveys (August 2001, September 2001, August 2002 and December 2003). This is not surprising given that i) authorised discharges of liquid effluent from the site to the sewer system are episodic and ii) the overflow of the sewer system to the CSO at Whitchurch Brook is intermittent.

A plot of the River Taff data to show temporal variation is provided in Fig. 3b. To a first approximation, activities remained essentially constant between November 2001 and December 2003, within the scatter of the data, with a median value of 3.7 Bq/kg. Somewhat greater activities were observed on the first two surveys. The scatter in the data is perhaps inevitable given that the volume and flow of water in the river is itself variable.



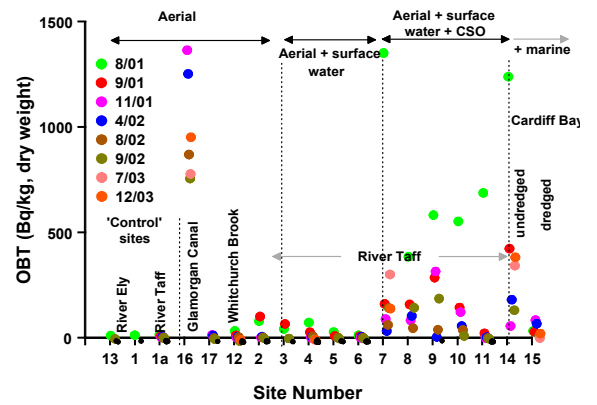
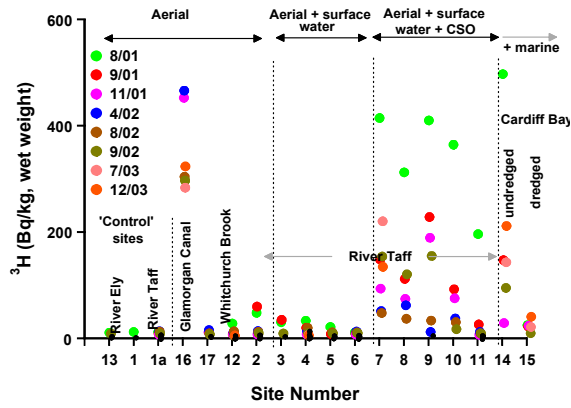
## 3.2 Sediments

### 3.2.1 *Tritium*

Results for  $^3\text{H}$  in sediment are given in Figs. 4a and 4b. Samples were analysed both wet and dry. The average  $^3\text{H}$  loss on drying was 15 %. The percentage of  $^3\text{H}$  present as OBT may be calculated by difference [OBT (%) = 100-Loss (%)]. The percentage of OBT was, therefore, ~85%. Due to the previously mentioned potential for OBT decomposition during the drying process, this value represents a lower limit for the fraction of  $^3\text{H}$  present as OBT. The different physical nature of the various sediment samples meant that the water content of individual samples was extremely variable, ranging from 19 % up to 81 % with an average value of 62 %. Consequently, when comparing values between individual sites, it is more useful to assess either the OBT or total  $^3\text{H}$  data that has been normalised to allow for the variable water content.

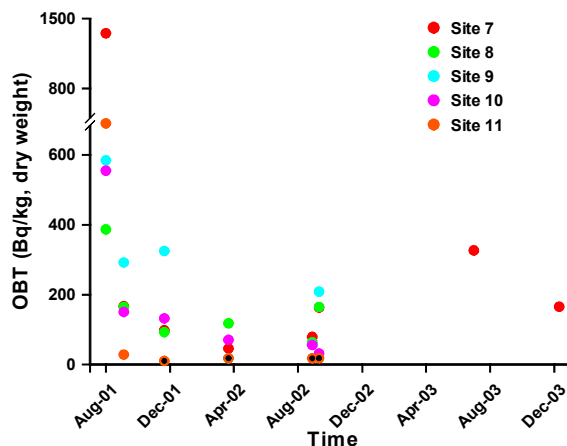
Figure 4

Tritium activity in sediment.



a) Wet sediment. Black dots indicate less than values.

b) Dry sediment (OBT). Black dots indicate less than values.



The OB T activity in sediments at the ‘control’ sites was below limit of detection (typically ~13 Bq/kg dry weight). The greatest activity (~1400 Bq/kg dry weight) was observed in a single sample from Glamorgan Canal (site 16), concomitant with that for dissolved  $^3\text{H}$  (Fig. 3a). In contrast to data for dissolved  $^3\text{H}$  in Whitchurch Brook (Fig. 3a, site 12), levels in sediments were low (average concentration of 14 Bq/kg dry

weight), and close to the limit of detection. Uptake of  $^3\text{H}$  by sediment may occur via physico-chemical sorption processes or bacterial transformation of dissolved organic  $^3\text{H}$  labelled compounds into particulate organic matter. The reason for the disparity in sediment activities between Whitchurch Brook and Glamorgan Canal could therefore be due to differences in sediment composition (grain-size and organic matter content) or bacterial activity between the two sites.



A total of 27 sediment samples have been analysed from sites 2-6 (i.e. River Taff upstream of the confluence with Whitchurch Brook) of which 19 were below limit of detection. The maximum concentration was observed in September 2001 at Site 2, close to the Amersham plc plant but upstream of the surface water run off. Samples collected downstream from site 2 on the same survey, but upstream of the confluence with Whitchurch Brook, contained lower OBT activities. In contrast, sediment collected in November 2001 from the site 2 contained levels of OBT below the detection limit. The difference may be a reflection of the varying mobility of sediment between low and high flow conditions of summer and winter, respectively. The average daily flow in the Taff is  $19.1 \text{ m}^3/\text{sec}$ , whereas the typical dry summer flow is  $3.5 \text{ m}^3/\text{sec}$  ([www.environment.agency.gov.uk/commondata/105385/taffov.pdf](http://www.environment.agency.gov.uk/commondata/105385/taffov.pdf)). It is therefore suggested that the most plausible explanation for the enhanced concentration at site 2 in September 2001 is episodic input to mobile sediments via atmospheric deposition. The data in Fig 4b indicate that the impact of site surface water run-off upon levels at site 3 and further downstream was minimal. Most importantly, levels at site 6, immediately upstream of the CSO, were below the detection limit in five out of six samples. This indicates

the influence of aerial deposition upon sediment activities in the River Taff is extremely small.



A total of 26 sediment samples have been analysed from sites 7-10 (i.e. River Taff downstream of the confluence with Whitchurch Brook and upstream of Blackweir). Only one sample was below the detection limit. Levels ranged from <math><18-1353\text{ Bq/kg}</math> dry weight. Despite the very large scatter in the data, it is apparent that OBT activities were noticeably elevated by the CSO input via Whitchurch Brook. A total of 6 sediment samples have been analysed from site 11, immediately downstream of Blackweir, of which 4 were below the detection limit. Only the first sample, collected in August 2001, contained a concentration comparable to those observed upstream of Blackweir at sites 7-10.



A plot of the River Taff sediment OBT data, downstream of the CSO (sites 7-11) to show temporal variation is provided in Fig. 4c. There was no overall trend in the data between September 2001 and December 2003. Significantly greater activities were, however, observed on the first survey. In addition to the differences in sediment mobility between different flow conditions, another contribution to the scatter in the OBT data may be burial of riverbed sediment by riverbank sediment. Indeed, work was being carried out on the riverbank when sampling was undertaken in August 2002, resulting in the presence of additional suspended sediment in the watercolumn.



Analyses were not been carried out on samples of suspended particulate material. This is because the weight of material available for analysis was very low and

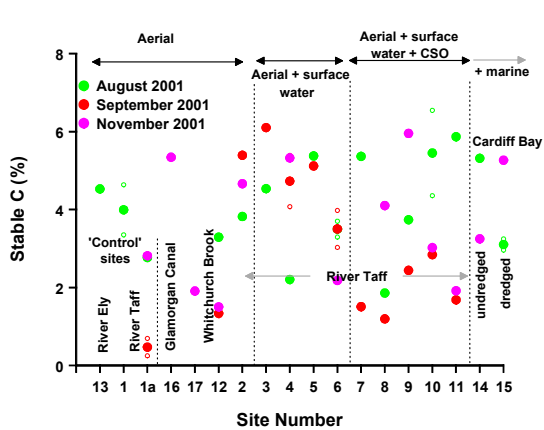
therefore the quantity of  $^3\text{H}$  in these samples would have been below the limit of detection.

### 3.2.2 Carbon-14

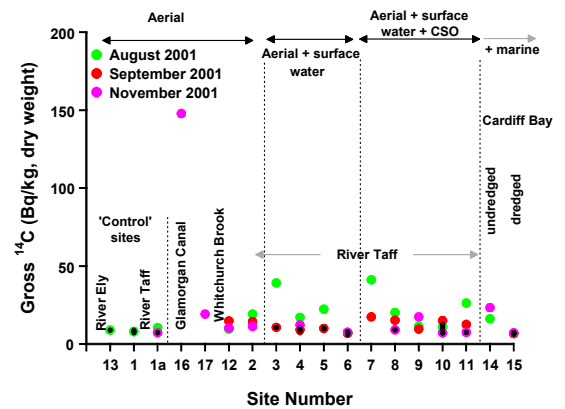
Results for the stable carbon content, gross  $^{14}\text{C}$  activity and  $^{14}\text{C}$  activity per kg stable carbon of sediment samples are given in Figs 5a, 5b and 5c, respectively.

Figure 5

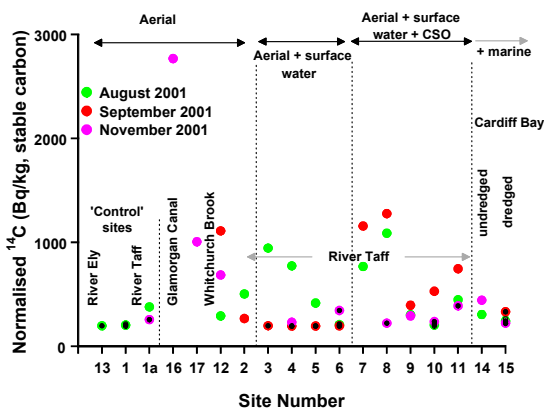
Stable carbon, and  $^{14}\text{C}$  activity, in sediment (August & September 2001).



a) Stable carbon content (%)



b) Gross  $^{14}\text{C}$  activity. Black dots indicate less than values



c) Normalised  $^{14}\text{C}$  activity (per kg stable carbon). Black dots indicate less than values.

The stable carbon content of the sediment samples ranged from 0.7-6.1 % with a median value of 3.5 % (Fig. 5a). The gross  $^{14}\text{C}$  content ranged from  $<8$  Bq/kg dry weight up to 148 Bq/kg dry weight, with a median value of  $\sim 11$  Bq/kg dry weight (Fig. 5b). Normalised to the stable carbon content (Fig. 5c), the  $^{14}\text{C}$  activity ranged from  $<220$  Bq/kg carbon up to  $\sim 2700$  Bq/kg carbon, with a median value of  $\sim 340$  Bq/kg carbon. These results show that the impact of the aerial discharges was readily apparent in Glamorgan Canal and Whitchurch Brook, but not in the River Taff. A small and extremely localised elevation immediately downstream of the CSO, as a result of liquid discharges, was noticeable in the River Taff.  $^{14}\text{C}$  is ubiquitous in the environment, even at sites remote from nuclear facilities. The estimated UK ‘background’ level in 2001 was  $\sim 240$  Bq/kg stable carbon (this value was derived from data reported by Olet et al., 1997 and Cook et al., 1998). Therefore, although the median  $^{14}\text{C}$  activity was greater than the ‘background’ level, a significant number of samples (30 %) contained activities that were below the detection limit and hence similar to, or possibly below, ‘background’. Selected samples were reanalysed to confirm these results. These data are also shown in Figs 5a-5c.

Duplicate analyses were carried out on selected samples. Where these were undertaken, the open symbols in Figs. 5a-5c indicate results from individual analyses and the solid symbols the average value. There were noticeable differences (up to 2.2 %) between duplicate analyses of the stable carbon content (Fig. 5a). The median difference between duplicate values was 0.7 %. The Taff catchment once supported an intensive coal mining industry and associated processes ([www.mtaa.co.uk/river\\_taffx.html](http://www.mtaa.co.uk/river_taffx.html)). As recently as the 1980s, widespread pollution

was caused by the deep mining industry throughout the catchment. The coal industry has now declined to the extent that only one deep mine is currently operational at Tower Colliery, Hirwaun in the headwaters of the River Cynon. Opencast coal mining currently occurs at Graweth Farm at Pentrebach in Merthyr Tydfil. The opencast operations and the remaining deep mine are far better controlled nowadays and the impact on water quality is much diminished. Consequently, the reason for the variation between individual stable carbon analyses is probably because of sample inhomogeneity, arising from the presence of coal particles as a legacy of the historic coal mining activity.

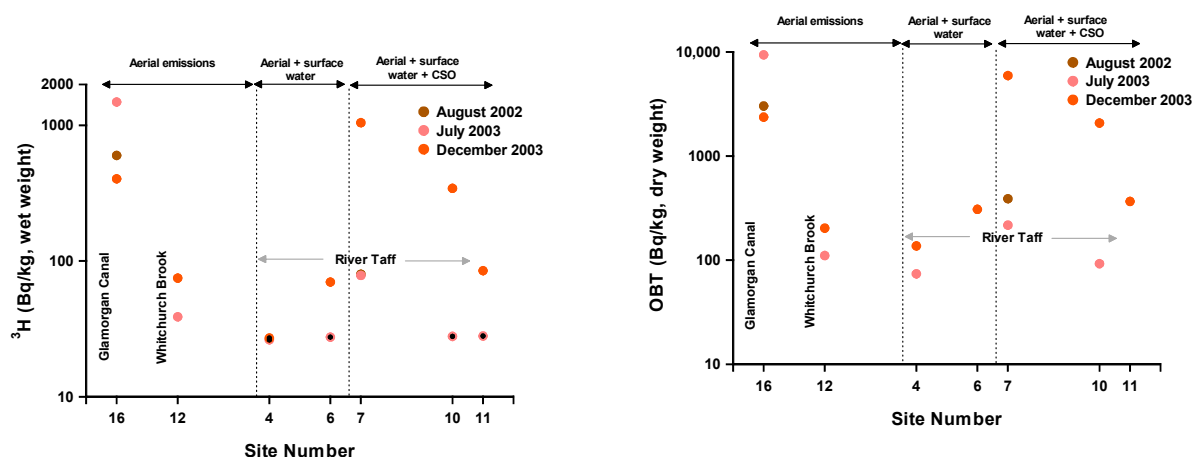
The replicate gross  $^{14}\text{C}$  data (Fig. 5b) and  $^{14}\text{C}$  activity normalised to the stable carbon content (Fig. 5c) were broadly consistent with the initial analyses in that the differences were relatively small, taking into account the uncertainty associated with individual analyses. Of equal importance is the fact that the data in Fig. 5c also confirm the initial findings that a number of sediment samples contain activity similar to, or possibly below, the estimated UK ‘background’ level in 2001. This reason for this can also be plausibly accounted for in terms of the presence of coal particles in the sediment samples. Coal particles, which because of their age contain no  $^{14}\text{C}$ , dilute the  $^{14}\text{C}$  activity in the sediment sample when it is normalised to the stable carbon content. This dilution is analogous to that of the perturbation of natural  $^{14}\text{C}$  to stable carbon ratios in the atmosphere produced by the burning of fossil fuels (the so called Suess effect).

### 3.3 Vegetation

Information concerning levels in vegetation is limited since sampling was confined to latter surveys (from August 2002) and vegetation was not always available. The vegetation consisted of riverweed from the River Taff and reeds from Glamorgan Canal. Analyses were restricted to  $^3\text{H}$  assay. The results are given in Figs. 6a and 6b. As for sedimentary material, samples were analysed both wet and dry. The average  $^3\text{H}$  loss on drying was 37 % and the percentage of OBT was, therefore, >63 %. The water content of individual samples ranged from ~83 % up to ~92 % with an average value of ~88 %. The variation in water content was considerably less than that observed for sediments.

Figure 6

Tritium activity in vegetation



a) Wet vegetation. Black dots indicate less than values.

b) Dry vegetation (OBT). Black dots indicate less than values.

Weed from sites in the River Taff (4-6) and Whitchurch Brook (12), primarily affected by aerial emissions, contained levels of total  $^3\text{H}$  in the range  $<30 - 75 \text{ Bq/kg}$  wet weight. At sites in the River Taff affected by the CSO (7-11),  $^3\text{H}$  activities ranged from  $<30-1050 \text{ Bq/kg}$  wet weight. Levels at site 7 immediately downstream of the CSO overflow were consistently elevated above those upstream.  $^3\text{H}$  activities in reeds collected from Glamorgan Canal (adjacent to the site) were also above background (range  $\sim 400-1490 \text{ Bq/kg}$  wet weight).

The concept of an equilibrium, biological concentration factor (CF) for vegetation and biota which indicates the apparent assimilation of radionuclides from aqueous solution has gained wide acceptance as a pragmatic measure despite the problems inherent in deriving and interpreting CF values (Lowman et al., 1971; IAEA, 1985). CF values are normally obtained from environmental observations whereby:

$$\text{CF} = \frac{\text{Concentration in biological material (Bq/kg, wet weight)}}{\text{Concentration in dissolved phase (Bq/kg)}}$$

Apparent CF values for weed and reeds were in the range 3-194 and 15-56, respectively. Given the scatter in the data for dissolved  $^3\text{H}$  activity (hence CF values) extreme caution is required in the interpretation and application (e.g. for modelling purposes) of these CF data.



### 3.4 Invertebrates

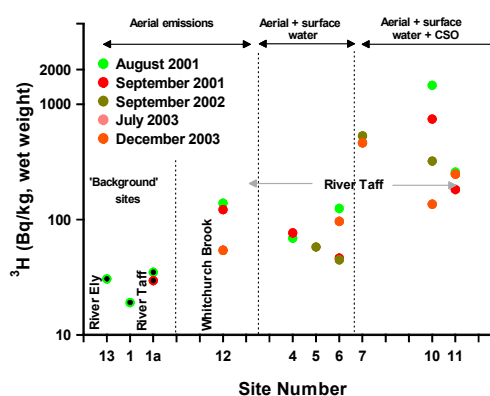
Assay of invertebrates (Fig. 7a) was restricted to  $^3\text{H}$ , given the limited quantity of material available for analysis. The results are given in Figs. 7b and 7c. The average  $^3\text{H}$  loss on drying was 6 % and the percentage of OBT was, therefore, >94 %. The water content of individual samples ranged from ~72 % up to ~92 % with an average value of ~86 %. Although variable, the range was less than that observed for sediments.

Figure 7

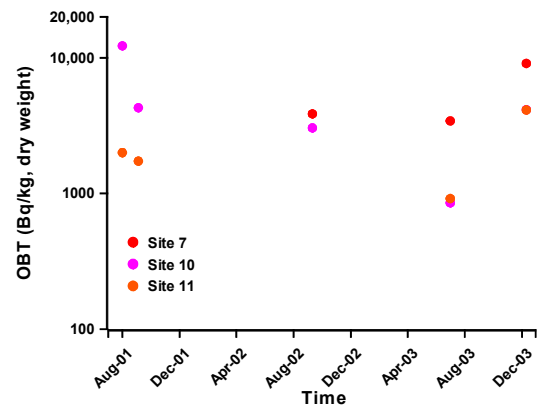
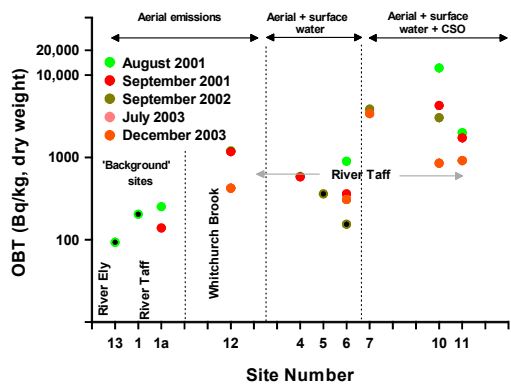
Tritium activity in invertebrates.



a) Picture of typical sample



b) Wet invertebrates. Black dots indicate less than values.



c) Dry invertebrates (OB T). Black dots indicate less than values.

d) Temporal variation in OB T activity. Samples were obtained from the River Taff downstream of confluence with Whitchurch Brook (sites 7,10 and 11).

Levels of both total  $^3\text{H}$  and OB T at the ‘control’ sites were below limit of detection (typically  $\sim 30$  Bq/kg wet weight and  $\sim 140$  Bq/kg dry weight, respectively). Sites in the River Taff (4-6) and Whitchurch Brook (12), solely affected by aerial emissions, contained levels of  $^3\text{H}$  that were measurably above the limit of detection. Activities at sites 4 -6 ranged from  $\sim 45$  -125 Bq/kg wet weight, with an average value of  $\sim 74$  Bq/kg wet weight. Levels at site 12 were broadly similar, ranging from  $\sim 54$  -139 Bq/kg wet weight, with an average value of  $\sim 110$  Bq/kg wet weight. The apparent CF values in the River Taff (upstream of the CSO) and Whitchurch Brook were in the range 7-27 and 4-8, respectively.

At sites in the River Taff affected by the CSO (7-11),  $^3\text{H}$  activities ranged from  $\sim 140$ -1730 Bq/kg wet weight with an average value of  $\sim 710$  Bq/kg, wet weight. Despite the low  $^3\text{H}$  activities in sediment downstream of Blackweir (site 11), it is notable that measurable amounts of  $^3\text{H}$  were observed in all three invertebrate samples. Pooling the data for sites 7, 10 and 11, indicates that the average concentration was elevated

by ~10 fold compared with that observed upstream of the CSO. Apparent CF values at sites 7-11 were in the range 26-406.

As mentioned previously, the average water content of the invertebrate samples was 86 %. This implies the average dry solids content was 14 %, which is roughly four fold less than that of the sediment samples (62 %). It is, therefore, only possible to compare levels of  $^3\text{H}$  in invertebrates and sediments for dry material (i.e. OBT values). OBT activities in invertebrates at all sites (Fig 7c) were markedly greater (typically between one and two orders magnitude) compared with those observed in the sediments (Fig. 4b). It is suggested that the  $^3\text{H}$  assimilation occurs primarily as a result of the invertebrates feeding on tritiated particulate organic matter/sediment dwelling microbes.

A plot of the River Taff invertebrate OBT data, downstream of the CSO (sites 7, 10 and 11), to show temporal variation is provided in Fig. 7d. It is not possible to identify any increase or decrease with time, due to the scatter in the data.

Finally, it is worth noting that, despite the logistical problems involved in the collection of sufficient quantities of invertebrates for  $^3\text{H}$  analysis, they probably constitute a better indicator material than sediments for routine monitoring.

## 3.5 Fish

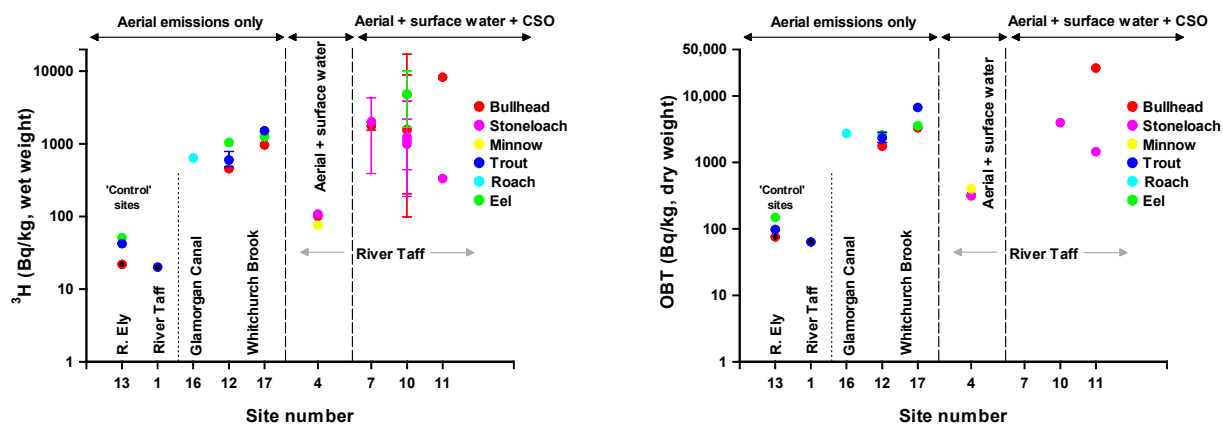
### *3.5.1 Bulk samples*

#### *3.5.1.1 Tritium*

The prime focus of this study was the impact of discharges upon radionuclide activities in fish. A large number of analyses have been carried out both on individual fish and bulk samples. As mentioned previously, it was not possible to obtain samples of all species from each site. Moreover, the size distribution (and by implication the age) in samples of small fish species was variable between individual sites. Data for bulk samples, together with average values from analyses of individual fish from surveys in 2001, are shown in Figs. 8a and 8b, to enable an initial assessment of the impact of aerial and liquid  $^3\text{H}$  inputs. Bulk samples were analysed both wet and dry. The average  $^3\text{H}$  loss on drying was 8 % and the percentage of OBT was, therefore, >92 %. The water content of individual samples ranged from ~61 % up to ~79 % with an average value of ~74 %.

Figure 8

Tritium activity in bulk fish samples (August and September 2001).



a) Wet fish. Black dots indicate less than values. Results for some samples estimated by calculating average value from analyses of individual fish. Error bars indicate maximum and minimum values

b) Dry fish (OBT). Black dots indicate less than values.

The results in Fig. 8a indicate  $^3\text{H}$  in fish from the ‘control’ sites was below the limit of detection ( $\sim 20$  Bq/kg wet weight), with the exception of a single trout sample from the River Ely (concentration  $\sim 40$  Bq/kg, wet weight).

Fish obtained from the River Taff downstream of Radyr Weir (site 4), close to the Amersham plc plant, contained  $^3\text{H}$  activities that were measurably above the detection limit ( $\sim 100$  Bq/kg wet weight). The apparent CF from filtered water was in the range 17-23. Fish from Whitchurch Brook (sites 12 and 17) and Glamorgan Canal (site 16), with levels of dissolved  $^3\text{H}$  markedly elevated by aerial deposition, contained  $^3\text{H}$  activities in the range  $\sim 460$ - $1520$  Bq/kg wet weight with an average concentration of  $\sim 930$  Bq/kg wet weight. The  $^3\text{H}$  concentration ( $640$  Bq/kg wet weight) in roach from Glamorgan Canal (site 16), indicated levels were similar to those in Whitchurch

Brook. The apparent CF from filtered water was in the range 26-60, with average value of ~40. These results indicate significant  $^3\text{H}$  assimilation may occur at sites affected solely by aerial emissions.

$^3\text{H}$  assimilation in bullheads and stone loach obtained from the River Taff downstream of the confluence with Whitchurch Brook (sites 7-11) ranged from ~330 Bq/kg wet weight up to 8300 Bq/kg wet weight. The average value (~2700 Bq/kg wet weight) was greater by ~30 fold compared with those observed at Radyr weir (site 4) upstream of the CSO input and of similar magnitude to that previously noted for invertebrates at site 7. In contrast to the relatively small range of  $^3\text{H}$  activities in fish from Whitchurch Brook, the variability in samples from sites 7-11 in the Taff was large (ranges varied by ~3 and 30 fold, respectively). The apparent CF was in range 48-1380 with average value of ~460. Closer inspection of the data in Fig. 8a shows that only a few samples contained levels approaching those observed in the August 2000 survey (7800-30,000 Bq/kg wet weight, Fig.1).

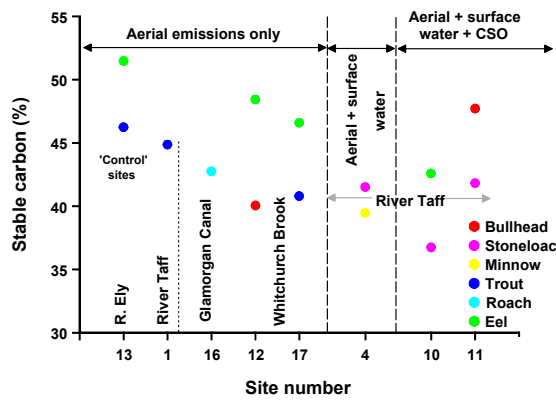
The assimilation of  $^3\text{H}$  by fish presumably occurs as a result of feeding on  $^3\text{H}$  labelled invertebrates (see discussion section). However, the dry solids contents of the fish (average was 26 %) was roughly double that of the invertebrates (average of 13 %) and less than half that of the sediments (average of 62 %). It is, therefore, only possible to compare levels of  $^3\text{H}$  in fish, invertebrates and sediments for dry material (i.e. OBT values). OBT activities in the fish at all sites were markedly greater (up to almost two orders magnitude) than those observed in the sediments. The ratio between the concentration in fish and the average value in invertebrates at a given site varied between 0.5 and 14 with an average value of 3.

### 3.5.1.2 Carbon-14

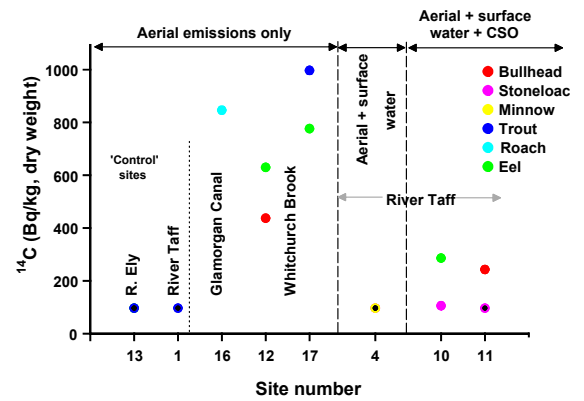
Results for the stable carbon content, gross  $^{14}\text{C}$  and  $^{14}\text{C}$  concentration per kg stable carbon of bulk fish samples are given in Figs 9a, 9b and 9c, respectively. In addition, a log-log plot of  $^{14}\text{C}$  versus  $^3\text{H}$  activities is provided in Fig. 9d.

Figure 9

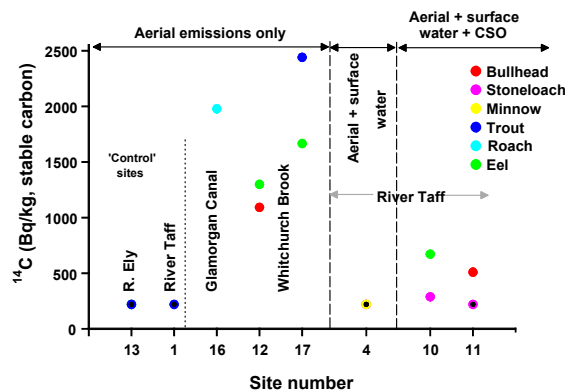
Stable carbon, and  $^{14}\text{C}$  activity, in fish from River Taff (August and September 2001).



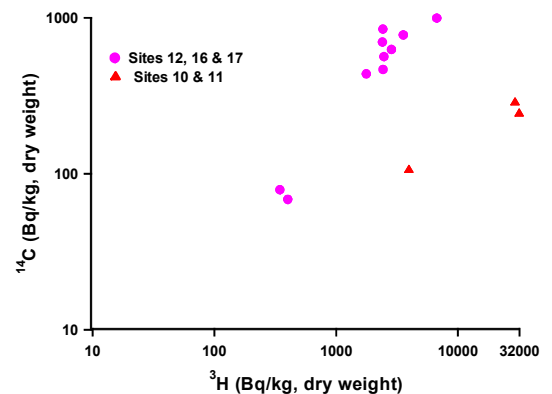
a) Stable carbon content.



b) Gross  $^{14}\text{C}$  activity. Black dots indicate less than values.



c) Normalised  $^{14}\text{C}$  activity (per kg stable carbon). Black dots indicate less than values.



d) log-log plot of gross  $^{14}\text{C}$  activity versus  $^3\text{H}$ .

The data in Fig. 9a indicate that the stable carbon content of the fish samples ranged from 37-54 % with a median value of 45 %. Proteins, which are the condensation products of up to 20 amino acids, occur in varying amounts in most food products. In the dry tissue of fish muscle the content of proteins is nearly 90% (Kirchmann et al., 1979). The great variance in quantitative and qualitative composition makes it difficult to estimate theoretically the hydrogen content and the percentage of nonexchangeable hydrogen. If, however, it is assumed that all 20 amino acids occur in equal proportions then the theoretical carbon content of protein is ~46%. The results for the stable carbon in Fig. 9a are, therefore, broadly consistent with the theoretical predictions.

The gross  $^{14}\text{C}$  activity (Fig. 9b) ranged from <100 Bq/kg dry weight up to 997 Bq/kg dry weight, with a median value of 176 Bq/kg dry weight. Normalised to the stable carbon content (Fig. 9c), the  $^{14}\text{C}$  activity ranged from <220 Bq/kg carbon up to 2440 Bq/kg carbon, with a median value of 400 Bq/kg carbon. Activities below our detection limit (~220 Bq/kg carbon), and possibly slightly lower than the estimated 2001 UK background level of ~240 Bq/kg carbon, were observed in samples from both 'control' sites (1 and 13) and in the River Taff at site 4 (upstream of the CSO overflow).

With respect to the influence of liquid discharges (fish from Taff downstream of the confluence with Whitchurch Brook), data are available for samples of Stoneloach, Bullheads and Eel from Sites 10 and 11.  $^{14}\text{C}$  activities in the stoneloach were close to UK 'background' level with activities in the range <220-288 Bq/kg stable carbon (Fig. 9c). In contrast, activities in fillet material from the Bullhead and Eel muscle

were noticeably elevated (by 2 –3 fold) above ‘background’. Therefore, the impact of the liquid discharges was small.

Significantly higher  $^{14}\text{C}$  activities in fish were in samples from Whitchurch Brook and the Glamorgan Canal. Levels in fish obtained at site 12 (Whitchurch Brook at the Three Elms Pub) were in the range ~1010-1340 Bq/kg stable carbon. The median value value (~1260 Bq/kg stable carbon) was ~5 fold greater than the background value. Even higher levels were observed further stream in Whitchurch Brook at site 17 (activities in the range 1670-2440 Bq/kg stable carbon). The  $^{14}\text{C}$  concentration observed in a single Roach from Glamorgan canal (site 16) was 1980 Bq/kg stable carbon (i.e. very similar to that in the fish from site 17). It is apparent from these data that the impact of the gaseous  $^{14}\text{C}$  discharges upon levels in fish is significantly greater than the liquid discharges. In turn, this is probably due to the fact that  $^{14}\text{C}$  dilution is greater in the fast flowing Taff compared with Whitchurch Brook and the Glamorgan Canal.

A log-log plot of gross  $^{14}\text{C}$  activity versus OBT (Fig. 9d) illustrates the degree of correspondence between assimilation of these two radionuclides. At sites 12, 16 and 17 (impacted by aerial discharges only), there is a linear relationship ( $r = 0.86$ ) between the activities as described by the regression line:

$$\log ^{14}\text{C} = (0.15\pm 0.03)*\log ^3\text{H} + 181\pm 94.$$

Although there was also a linear relationship ( $r = 0.97$ ) between  $^{14}\text{C}$  and  $^3\text{H}$  activities for fish obtained from sites 10 and 11 in the river Taff (impacted by liquid discharges via the CSO from Whitchurch Brook), the slope and intercept were different to those for sites 12, 16 and 17.  $^{14}\text{C}$  and  $^3\text{H}$  activities fitted to the regression line:

$$\log {}^{14}\text{C} = (0.006 \pm 0.001) * \log {}^3\text{H} + 86 \pm 23$$

### 3.5.2 Tritium activities in individual fish

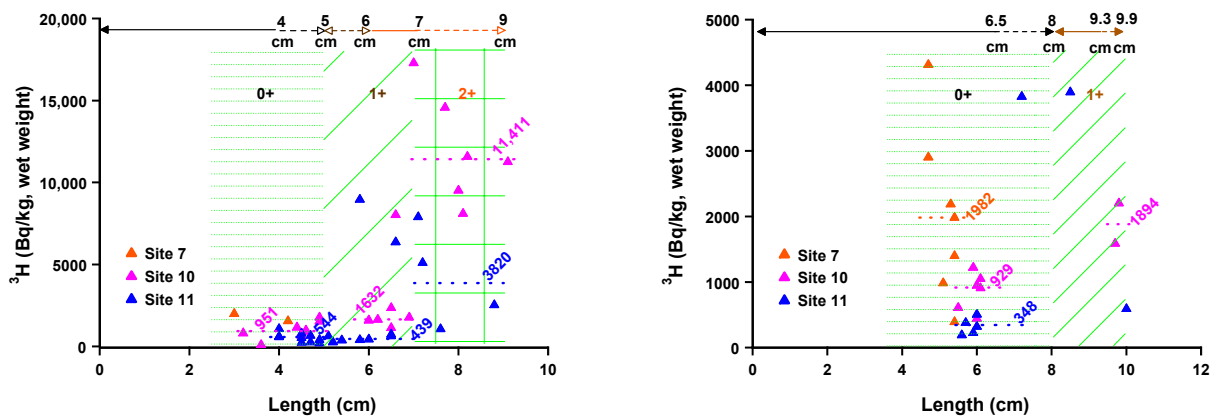
The data in Figs. 8a and 8b highlighted some of the variability in  ${}^3\text{H}$  activities between individual bulk fish samples, particularly from sites 7-11. Selective analyses of individual fish were undertaken in an attempt to gain a better understanding of reasons for the differences.

#### 3.5.2.1 2001 survey data

Results for analyses of total  ${}^3\text{H}$  activity in individual fish from surveys in 2001, as a function of fish length, are provided in Figs. 10a-10d. Dotted lines and associated text have been included to indicate median values for fish over specified length ranges, along with information to show typical ranges in length for individual age groups (arrows).

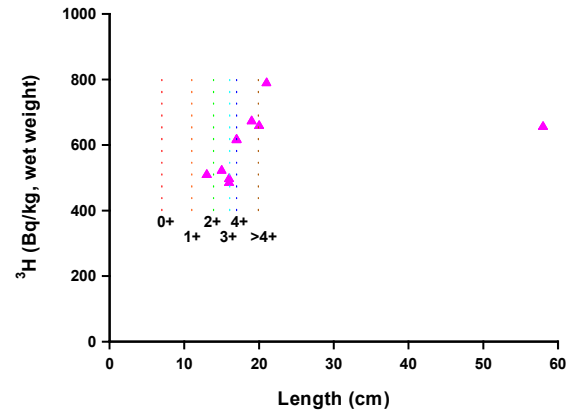
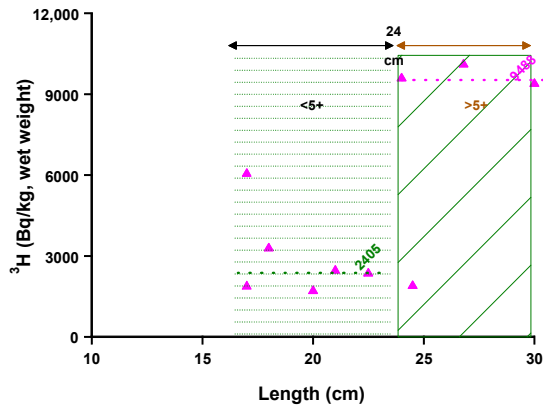
Figure 10

Variation in tritium activity between individual fish obtained from Whitchurch Brook and the River Taff downstream of CSO (August and September 2001).



a) Bullheads from sites 7, 10 and 11. Dotted, shaded and cross-hatched areas indicate inferred 0+, 1 + and 2+ age groups.

b) Stone loach from sites 7, 10 and 11. Dotted and shaded areas indicate inferred 0+ and 1 + age groups.



c) Eels from site 10. Information concerning eel age inferred from length-age data for eels from Whitchurch Brook (Site 12). Dotted and shaded areas indicate inferred <5+ and >5+ age groups.

d) Trout from site 12. Dotted lines indicate mean length for different ages groups of trout from the Afon Cwm, mid-Wales (Crisp and Beaumont, 1995)

The total  $^3\text{H}$  activity in bullheads ranged from  $\sim 100 - 17,300$  Bq/kg wet weight and  $\sim 210 - 8960$  Bq/kg wet weight at sites 10 and 11, respectively (Fig. 10a). At both sites, the data indicate i) extremely large (up to two orders of magnitude) variation in levels between individual fish and ii) a noticeable discontinuity in  $^3\text{H}$  activities between the smallest (<5 cm) and largest (>7 cm) fish.

At site 10, levels in small fish with length <5 cm varied from 99-1765 Bq/kg wet weight (i.e. by  $\sim 18$  fold) with a median value of 951 Bq/kg wet weight (n=8). If the lowest value is excluded then the variation is reduced to just 3 fold. This outlying value could conceivably be a result of the migration downstream of a fish previously living upstream of the CSO. In contrast, activities in large fish of length >7 cm ranged from 8101-17,286 Bq/kg wet weight (i.e. variation of  $\sim 2$  fold) with a median value of 11,411 Bq/kg wet weight (n=6). The increase in median activities for >7 cm fish, compared with those of length <5 cm was  $\sim 12$  fold and was statistically significant ( $p < 0.01$ ). Data are also available for six intermediate size fish (lengths

between 6-6.9 cm, and covered by the shaded area in Fig. 10a). Levels were in the range 1134-8017 Bq/kg wet weight (i.e. variation of ~ 7 fold). The scatter of the data over this range of length complicates the selection of a unique cut-off point for the  $^3\text{H}$  discontinuity. It is worth noting that just one out of the six fish (length 6.6 cm) contained a  $^3\text{H}$  concentration characteristic of the larger (>7 cm) fish. Excluding this single high outlying value, the range is reduced to 1134-2343 Bq/kg wet weight (i.e. variation of just ~ 2 fold) with a median value of 1632 Bq/kg weight (n=5).

At site 11, levels in the small fish with length in the range 4.0-4.9 cm varied from 205-1073 Bq/kg wet weight (i.e. by ~5 fold) with a median value of 544 Bq/kg wet weight (n=10). In contrast activities in large fish of length >7 cm ranged from 1059-7883 Bq/kg wet weight (variation of ~7.4 fold) with a median value of 3820 Bq/kg wet weight (n=4). The increase in median activities for >7 cm fish, compared with those of length <5 cm was ~ 7 fold and was statistically significant ( $p < 0.01$ ). In addition, there was a noticeable difference in median activities between sites 10 and 11 (levels greater at site 10 by ~1.7 fold and 3.0 fold, respectively, for <5 cm and > 7 cm fish). The difference was statistically significant at the 5 % confidence level ( $p < 0.02$ ). Data are also available for nine intermediate size fish from site 11 (lengths between 5.2-6.6 cm, and covered by the shaded area in Fig. 8a). Levels were in the range 258-8960 Bq/kg wet weight (i.e. variation of ~ 35 fold). The scatter of the data was even greater than that observed at site 10, and tends to mask the  $^3\text{H}$  discontinuity between small and large fish. Two out of the nine fish (those of length 5.8 cm and 6.6 cm) contained  $^3\text{H}$  activities characteristic of the larger (>7 cm) fish. The range of values in the remaining seven was reduced to 258-739 Bq/kg wet weight (i.e. variation of just ~ 3 fold) with a median value of 439 Bq/kg weight (n=7).

Information is also provided in Fig. 10a, to illustrate typical variations in fish length with age. Generally, bullheads attain a length of 4–5 cm after their first year, 6 cm after their second and 7–9 cm after their third (Maitland and Campbell, 1992). Fish with length < 4 cm were, therefore, almost certainly from the 0 age group and spawned in 2001. However, the growth rate, and other parameters such as sexual maturity and longevity, varies significantly between different environments. For example, in the productive chalk streams of southern England individuals grow rapidly and attain lengths of 5-6 cm after one year (Fox, 1978, Mills and Mann, 1983). In the absence of specific information for the River Taff it is, therefore, impossible to accurately relate fish length and age. Nevertheless, even assuming higher than typical growth rate, bullheads whose length was greater than 8 cm were likely to be more than 2 years old. Given that bullheads do not generally live for more than three or four years, these fish were probably spawned between 1997-1999. Combining the information in Fig. 8a concerning the variations in  $^3\text{H}$  concentration and age with length it is apparent that 0+ age group fish spawned in 2001 contained substantially lower activities than the largest (> 8 cm) fish that were probably more than two years old. In addition, it is tentatively suggested that the median values identified for < 5 cm, 5-7 cm and > 7 cm fish represent those for 0+, 1+ and 2+ age groups. The three outlying values for 5-7 cm fish, excluded when calculating median values, are suggested to be small 2+ age group fish resulting from natural variation in growth rate.

Total  $^3\text{H}$  activities in stone loach ranged from 391 – 4312 Bq/kg wet weight, 440 – 2203 Bq/kg wet weight and ~190 – 3896 Bq/kg wet weight at sites 7, 10 and 11, respectively (Fig. 10b). The variation in levels between individual fish was, therefore,

in the range 5-21 fold. Information concerning the growth of stone loach in the Lake District indicates the mean length at the end of each year of life was: 8 cm in the first year; 9.9 cm in the second year and 10.8 cm in the third year (Smyly, 1955; Wheeler, 1969). Somewhat lower growth rates have been reported for a highly productive chalk stream in Dorset (Mills et al., 1983). Mean lengths after 1 and 2 years were 6.5 cm and 9.3 cm, respectively. Once again, in the absence of specific information for the River Taff it is, impossible to accurately correlate fish length with age. Nineteen out of the twenty three fish analysed here had length < 8 cm and could have been spawned in 2001. Given the lack of > 8 cm stone loach obtained in this survey, it was not possible to analyse enough samples to assess whether there was a significant difference in activities between < 8 cm and > 8 cm fish. Median activities in <8 cm fish were 1982 Bq/kg wet weight (n=7), 929 Bq/kg wet weight (n=5) and 348 Bq/kg wet weight (n=6) at sites 7, 10 and 11, respectively and decrease with distance from the CSO. The difference between levels at sites 7 and 10 was statistically significant at the 5 % confidence level. In contrast, the difference between results for sites 10 and 11 was not significant in statistical terms due to the high concentration found in one 7.2 cm fish obtained from site 11. This outlier could be an undersized 1+ fish. Finally, it is worth noting that statistical analysis indicated median values for < 8 cm (possibly 0+ age group) stone loach at individual sites were similar to those for < 5 cm (0+) bullheads.

Total <sup>3</sup>H activities in site 10 eels ranged from 1705 – 10,102 Bq/kg wet weight (Fig. 10c). The variation in levels between individual eels was, therefore, ~6 fold.

Information concerning the length-age relationship of the eels analysed here was inferred from data for the Whitchurch Brook tributary. Eels collected from Site 12

had lengths ranging from 18-45 cm. Their ages were determined to be in the range 3+ - 14+ years (SE Wales Environment Agency Appraisal Team, per comm.). A plot of Age versus length indicated the data fitted well ( $r=0.964$ ) to a linear regression line of the form:

$$\text{Age} = (0.36 \pm 0.03) * \text{length} - (3.5 \pm 0.1)$$

Application of this relationship to site 10 eels whose length ranged from 17-30 cm indicated their ages varied from ~3+ - 7+ years (Fig. 10c). It is notable that the median  $^3\text{H}$  concentration in eels with length (<24 cm), and hence age 3+ - 4+ years, was 2405 Bq/kg wet weight ( $n=6$ ) and similar to that in 1+ bullheads. In contrast the median activities in eels with length (>24 cm), and hence age 5+ - 7+ years, was 9488 Bq/kg wet weight ( $n=4$ ) which was greater by ~ 4 fold compared to that observed in younger animals. The difference was statistically significant at the 5 % confidence level ( $p<0.05$ ).

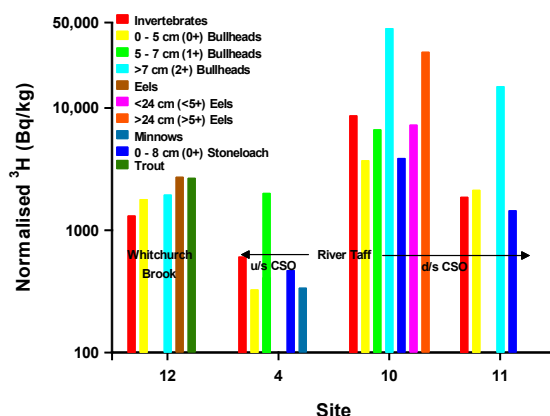
Total  $^3\text{H}$  activities in site 12 trout ranged from 485 – 789 Bq/kg wet weight (Fig. 10d) with an average value of  $602 \pm 98$  Bq/kg wet weight ( $n=10$ ). The variation in levels between individual fish was ~1.6 fold. This was considerably less than that observed at sites 7-11 for fish from the River Taff. Information concerning the growth of trout in the Afon Cwm (Crisp and Beaumont, 1995) indicates the mean length at the end of each year of life was: 14 cm in the second year; 16 cm in the third year, 17 cm in the fourth year and 20 cm for older fish. Application of these data to site 12 trout whose length ranged from 13-58 cm indicated their ages varied from ~1+ - >4+ years (Fig.

10d). It is noticeable that there is an apparent increase in  $^3\text{H}$  concentration with age between 2+ and 4+ years. It is, however, uncertain whether the increase is significant.

It is interesting to compare the results for individual fish age groups with those for invertebrates, after normalising the data to account for variable dry solids content. For example, levels in the two invertebrate samples from site 11 in 2001 were 258 Bq/kg wet weight (dry solids content of 11.7 %) and 182 Bq/kg wet weight (dry solids content of 11.9 %). Normalised to a dry solids content of 100 % these values are equivalent to ~2200 and ~1524 Bq/kg, respectively with a median value of 1862 Bq/kg. The median total  $^3\text{H}$  activities in < 5 cm bullheads from site 11 was 544 Bq/kg wet weight (dry solids content of 25.7 %). Normalised to a dry solids content of 100 % this is equivalent to ~2119 Bq/kg (i.e. very similar to that for the invertebrates). Median values of the normalised total  $^3\text{H}$  activity for fish and invertebrates from sites 12, 4, 10 and 11 are provided in Fig. 11.

Figure 11

Median values for total tritium activity in invertebrates and fish normalised to dry solids content of 100 % (August and September 2001).



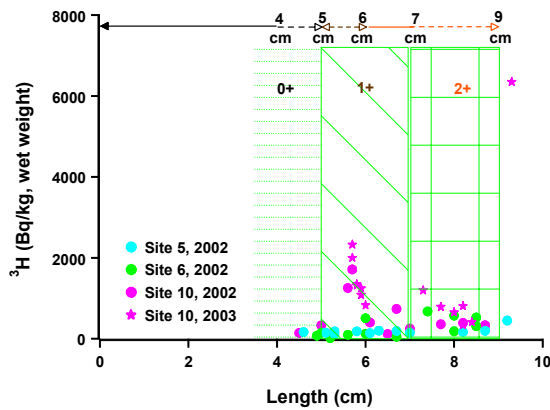
To a first approximation, median activities in fish were similar to those in invertebrates at individual sites. The outstanding exceptions are the data for i) >7 cm (>2+) bullheads at sites 10 and 11, where the normalised <sup>3</sup>H in fish was greater by 5 and 8 fold, respectively, compared to those in invertebrates and ii) >24 cm (>5+) eels at site 10 (normalised <sup>3</sup>H in eels greater by ~ 3 fold).

### 3.5.2.2 2002 and 2003 survey data

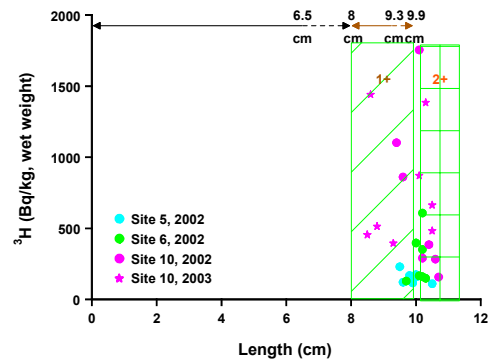
Results for analyses of total <sup>3</sup>H in individual fish from surveys in September 2002 and July 2003, as a function of fish length, are provided in Figs. 12a-12d. Information is also provided here to show median values for fish over specified length ranges (dotted lines and associated text) and typical ranges in length for individual age groups (arrows).

Figure 12

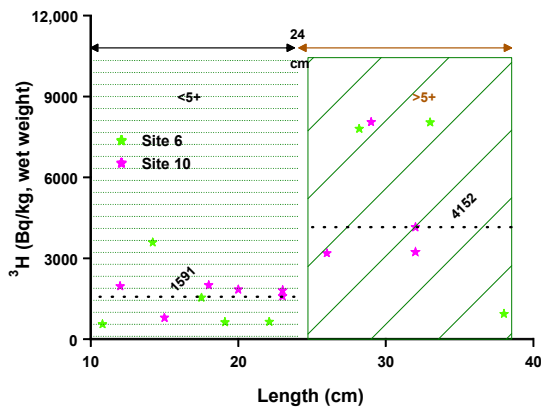
Variation in tritium activity between individual fish obtained from Whitchurch Brook and the River Taff downstream of CSO (September 2002 and July 2003).



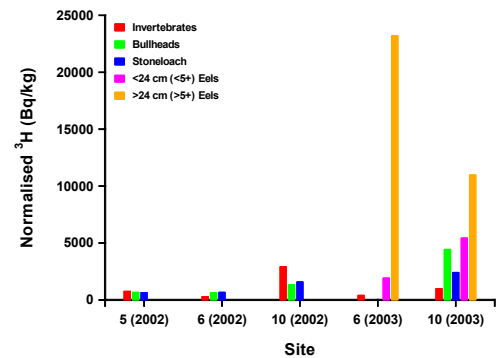
a) Bullheads from sites 5, 6 and 10. Dotted, shaded and cross-hatched areas indicate inferred 0+, 1 + and 2+ age groups.



b) Stone loach from sites 5, 6 and 10. Shaded and cross-hatched areas indicate inferred 1+ and 2 + age groups.



c) Eels from sites 6 and 10. Information concerning eel age inferred from length-age data for eels from Whitchurch Brook collected in August 2001 (Site 12). Dotted and shaded areas indicate inferred <5+ and >5+ age groups.



d) Median values for total tritium activities in invertebrates and fish normalised to dry solids content of 100 %

The results in Fig. 12a for total  $^3\text{H}$  activities in bullheads in 2002 indicate that, upstream of the CSO activities remained below 700 Bq/kg wet weight. Levels at sites 5 and 6 ranged from ~114– 449 Bq/kg wet weight and <83 – 677 Bq/kg wet weight, respectively. Downstream of the CSO at site 10, activities in some fish were notably enhanced ranging from 111 –1713 Bq/kg wet weight. With the notable exception of a single data point, in contrast to the data in Fig. 8a, there was no large increase in  $^3\text{H}$  activities between the smallest and largest fish. A full assessment is unfortunately limited by the small numbers of < 5 cm fish in the samples obtained in the 2002 and 2003 surveys.

Median bullhead activities were 175 Bq/kg wet weight (n=12), 166 Bq/kg wet weight (n=12) and 345 Bq/kg wet weight (n=12) at sites 5, 6 and 10, respectively. The difference in levels between sites 5 and 6 and 6 and 10 was not statistically significant. This is perhaps not surprising given the proximity of site 6 to the CSO (i.e. fish obtained from site 6 could migrate downstream of the CSO). There was, however, a statistically significant difference in median activities between sites 5 and 10 at the 95 % confidence level ( $p < 0.03$ ). The available data for 2003 indicate activities tended to be greater than those in 2002 (range 339-6348 Bq/kg wet weight with a median of 1141 Bq/kg, wet weight). The single very high value was found in a fish of length 9.3 cm. This fish could be in the >3+ or 4+ age group. The remaining fish were probably from the 1+-2+ age groups. Excluding this single high outlying value, the maximum activity was 2327 Bq/kg wet weight (i.e. similar range to that observed in 2002).

In contrast to the fish obtained in 2001 (Fig. 10b), all the stone loach obtained in the 2002 and 2003 surveys had length > 8 cm and were probably 1+ or 2+ fish (Fig. 12b). <sup>3</sup>H activities in 2002 ranged from ~111– 230 Bq/kg wet weight, 129 – 608 Bq/kg wet weight and 157-1754 Bq/kg wet weight at sites 5, 6 and 10 respectively. The median values (154 Bq/kg wet weight (n=6), 166 Bq/kg wet weight (n=6) and 385 Bq/kg wet weight (n=6) at sites 5, 6 and 10, respectively) were not statistically different to those for bullheads. Levels in stone loach at site 10 in 2003 (median value 589 Bq/kg wet weight, range 395-1441 Bq/kg wet weight) were remarkably similar to those observed in 2002.

<sup>3</sup>H activities in eels (Fig. 12c) tended to be greater than those observed in bullheads and stone loach. Levels at sites 6 and 10 ranged from ~552– 8037 Bq/kg wet weight and 792 – 8049 Bq/kg wet weight, respectively. As observed in 2001, there was a statistically significant difference between activities in < 24 cm and >24 cm eels. There was, however, no significant difference between the median values for individual length groups for the two sites. Pooling the data for the two sites yielded median activities of 1591 Bq/kg wet weight and 4152 Bq/kg wet weight of <24 cm and >24 cm eels, respectively. Levels in the older age group were, therefore, greater by ~ 3 fold compared to that observed in younger animals. An analysis has also been carried out to assess whether levels in at site 10 decreased between 2001 and 2003. The median concentration in < 24 cm eels in 2003 was lower by ~ 24 % compared to that observed in 2001. The decrease was significant at the 90 % confidence level. Although the median concentration in >24 cm eels in 2003 was less than half that observed in 2001, due to the scatter in the data the decrease was not statistically significant.

Median activities in bullheads and stone loach, normalised to account for variable dry solids content (Fig. 12d), were almost identical to those in invertebrates at site 5. Noticeable differences between fish and invertebrates, and between individual fish species/age groups, were observed at sites 6 and 10. Levels tended to be higher in fish. The greatest disparity was between >24 cm eels and invertebrates obtained during the 2003 survey (level in >24 cm eels higher by ~60 fold and ~ 11 fold and sites 6 and 10, respectively).

#### **4. Discussion**

Tritium and  $^{14}\text{C}$  are ubiquitous in the environment and have a variety of sources. They are both cosmogenic radionuclides being continuously produced in the upper atmosphere as a result of cosmic ray induced spallation and particle interactions with atmospheric nitrogen and oxygen. They are transported to the earth's surface via the hydrologic cycle. Levels have been further elevated by the legacy of atmospheric nuclear weapons testing between 1952 and 1962. Discharges to the aquatic environment from nuclear power and fuel reprocessing plants also produce locally enhanced activities. Consequently, the radionuclide activities in the samples analysed here are not solely due to releases from the Amersham plc plant.

#### 4.1 Tritium data

Comprehensive surveys of watercourses in the vicinity of the Amersham plc plant at Cardiff have been carried out since August 2001. To date, more than four hundred samples have been analysed for  $^3\text{H}$  and OBT. The information resulting from this programme has provided an indication of the impact of both aerial emissions and liquid discharges upon sediments and biota.

The levels of  $^3\text{H}$  in sediments, invertebrates and fish at the control sites were below the limit of detection. These sites were situated on the River Taff and River Ely some 3 km to the north and 8 km to the west of the Amersham plc plant, respectively. The short distance of these sites from the plant indicates the restricted area affected by the releases.

The aerial emissions did have a noticeable impact upon two small watercourses to the east of the Amersham plc plant. As expected, the greatest enhancement in levels of dissolved  $^3\text{H}$  was observed in Glamorgan Canal adjacent to the plant (distance <0.5 km). Levels found at sampling sites in Whitchurch Brook ~3 km from the plant were significantly lower. Aerial emissions had only a minor impact upon levels in the River Taff, due to a considerably greater dilution factor. Uptake by sediment, at these sites was variable. Activities were low and only just above the detection limit on coarse grained sandy material from Whitchurch Brook, but consistently high on fine-grained silty organic rich mud from Glamorgan Canal. In the River Taff, upstream of the CSO, activities in the majority of sediment samples were below the limit of detection. Measurable assimilation was found in invertebrates and fish (average CF

value for fish in Glamorgan Canal and Whitchurch Brook was 40). This finding was unexpected given that the aerial emissions consist largely of gaseous  $^3\text{H}_2$  and  $^3\text{H}_2\text{O}$  vapour. Results from laboratory studies to study the incorporation and distribution of  $^3\text{H}$  in aquatic organisms from exposure to  $^3\text{H}_2\text{O}$  do not indicate significant assimilation (Blaylock et al., 1986; Diabaté and Strack, 1993; Murphy, 1993).

The impact of liquid waste discharges, via the CSO, upon levels in water from the River Taff was barely detectable. In contrast, levels in sediment samples were clearly enhanced albeit that there was very large scatter in the total  $^3\text{H}$  and OBT data. The scatter limits application of sediment data as a monitoring tool. Assessment of the results for temporal variation did not indicate any overall trend between September 2001 and December 2003. Assimilation in invertebrates was clearly greater at sites impacted by discharges from the CSO (average concentration in the River Taff elevated by ~10 fold compared with that observed upstream of the CSO). Once again, to a first approximation, activities remained essentially constant throughout the study period. Levels of OBT at all sites were typically between one and two orders magnitude greater than those observed in the sediments.

The information concerning  $^3\text{H}$  in fish affected by CSO discharges was complex. Analyses of bulk samples from the initial surveys in 2001 (Fig. 8a) indicated an apparent decrease in levels compared with those observed in the August 2000 survey (Fig.1). Subsequent analyses of individual fish (Figs. 10a-10d) indicated significant variability in  $^3\text{H}$  activities, most notably in bullheads. Plots of  $^3\text{H}$  concentration versus fish length demonstrated a noticeable discontinuity in levels between the smallest and largest bullheads. An unequivocal interpretation of the data was

hampered by lack of specific information concerning increases in fish length concomitant with age in the River Taff. It is nevertheless suggested that the most plausible explanation for the observed behaviour is the presence of three separate age groups in the sample, containing different  $^3\text{H}$  activities. Median activities at site 10 in 2001 were ~950, 1630 and 11,410 Bq/kg wet weight in < 5 cm (0+), 5-7 cm (1+) and >7 cm (2+) bullheads. The extreme variability, due to the presence of bullheads spawned in or before 1999, was, with the exception of a single outlying value in 2003, absent in subsequent surveys (Fig. 12a). This is probably because bullheads are a short-lived species (typically 3-4 years).  $^3\text{H}$  activities in bullheads and stone loach, assumed to be in 0+ and 1+ classes, were similar at individual sites. However, in contrast to the bullhead data, results for stone loach in 2001 (Fig. 8b) did not show significant increase in  $^3\text{H}$  concentration concomitant with fish length. The most likely explanation is that the stone loach samples collected in 2001 did not contain fish from the 2+ age group.

The bullhead and the stone loach are generalist predators, whose diet is made up almost exclusively of benthic insects and crustaceans (Hyslop, 1982; Welton et al., 1983; Copp et al., 1994; Michel and Oberdorff, 1995). They adopt a flexible, opportunistic foraging strategy in response to fluxes of key invertebrate groups. They are also largely sedentary. In the eight-month study of Downhower et al. (1990), bullhead populations moved less than 50 m on average between captures. Since there is no reason for  $^3\text{H}$  to be biomagnified (Blaylock et al., 1986; Murphy, 1993), levels in bullheads and stone loach at a particular site would be expected to be similar to that in invertebrates (after normalising data to account for variable water content). The discrepancy between the data for >7 cm (>2+) bullheads and invertebrates at sites 10

and 11 in 2001 (Fig. 8) is, therefore, suggested to be a legacy of greater historic discharges from the CSO (i.e. in 1999 and preceding years).

Significantly variable activities, concomitant with length, were also observed in eels, in both 2001 and 2003 (Figs. 10d and 12c). It is perhaps fortuitous that it was possible to identify any differences from the analysis of this limited number of samples. This is because of the fact eels migrate over considerably greater distances than the sedentary small fish, allowing them to feed on material containing variable  $^3\text{H}$  activities. Median  $^3\text{H}$  concentration in eels with length ( $>24$  cm), and hence age  $<5+$  years, were greater by  $\sim 4$  fold, compared to those with length  $<24$  cm (i.e.  $<5+$  years). To interpret this behaviour, it is necessary to consider the life history of the eel. Spawning of leptocephali takes place in the western Atlantic. They then are then dispersed in the north-easterly moving ocean currents. Metamorphosis, from leptocephalus to a transparent eelshaped 'glass eel', also takes place in the ocean and they arrive on European coasts after  $2\frac{1}{2}$ -3 years. The glass eels undergo what has been called a second metamorphosis when pigment cells form in the skin and a fully coloured elver is formed. This second metamorphosis takes place in coastal waters. After metamorphosis the elvers ascend rivers or take to life in the littoral zone at a length of about 7 cm. Off the Channel, Irish Sea and Scottish coasts, this occurs in February. As with bullheads and stone loach, the eel's diet is composed almost entirely of bottom-living invertebrates, chiefly molluscs and insect. Crustaceans and fish are rarely eaten. The three year time lag between the  $^3\text{H}$  discontinuity in eels (at 5+ years, Fig. 10d) compared with the bullheads (at 2+ years, Fig. 10a) is, therefore, a result of the fact that eels live in the sea for their first 3 years of life.

Overall, the data presented here indicate that the impact of liquid wastes entering the River Taff from the CSO was markedly reduced in 2001 compared with that in 1999 and preceding years (as evidenced by the noticeable discontinuity in levels between the smallest and largest bullheads in 2001). However, the total  $^3\text{H}$  activity in the liquid discharges has only reduced slightly since 1999 (Table 1). Moreover, there has been no change in the operation of the CSO overflow in recent years albeit that the Cardiff sewer system is being upgraded (Welsh Water, pers. comm.) to the sewage system. Finally, between May 1998 and April 2003 the chemical form (hence bioavailability) of  $^3\text{H}$  compounds present in the liquid waste remained broadly constant (Amersham plc, pers comm.). Although it is conceivable that, between the summer of 1999 and the spring of 2000  $^3\text{H}$  bearing riverbed sediment was buried beneath unlabelled riverbank sediment as a result of riverworks, the reason for the apparent decrease in  $^3\text{H}$  activities in biota after 1999 remains unknown. Whatever the most appropriate reason, the impact of liquid discharges entering the River Taff via the CSO overflow upon short-lived fish species such as bullheads is now relatively minor. Levels in some longer-lived species such as eels remain elevated in older fish, as a legacy of historically impact greater of CSO releases. As mentioned previously, in April 2003, the organic portion of the tritiated discharge was reduced by around 60%. Therefore, it is reasonable to anticipate a future trend of decreasing  $^3\text{H}$  activity in biota.

#### 4.2 Carbon-14 data

Assessment of the impact of  $^{14}\text{C}$  discharges upon levels in the materials analysed here is complicated. Given that the stable carbon content of natural materials varies widely,

measurements are usually quoted in units of Bq/kg stable carbon. From information published elsewhere (Otlet et al., 1997; Cook et al., 1998) concerning the  $^{14}\text{C}$  activity in materials at sites remote from nuclear facilities in the UK, the background level in 2001 was estimated to be ~240 Bq/kg stable carbon. This value is only slightly greater than the detection limit of the method used in the present study (~220 Bq/kg stable carbon). The  $^{14}\text{C}$  activity in a significant number of sediment samples, normalised to stable carbon content, was below the detection limit, hence the background level. It has been suggested that this is due to the presence of coal particles in the sediment (because of their age they do not contain  $^{14}\text{C}$ ) from the legacy of mining operations in South Wales. Based on the limited available data, the impact of the aerial discharges was readily detectable in Glamorgan Canal and Whitchurch Brook, but not in the River Taff. This is probably due to variations in water flow (hence dilution and dispersion) between the watercourse. The impact of the liquid discharges, via the CSO overflow, upon sediment in the River Taff was only apparent immediately downstream of the confluence with Whitchurch Brook.

Similar trends to that noted for sediment were apparent in data for fish. Levels in samples from the River Taff upstream of the CSO overflow were apparently below the background level. Work reported elsewhere indicates that it is possible that levels of  $^{14}\text{C}$  in freshwater fish may be depressed when accumulated terrestrial peat carbon is transferred to aquatic insect larvae and passed on to higher organisms (Schell, 1982). Levels were slightly elevated in some samples from the River Taff obtained downstream of the CSO overflow with more marked effects apparent in Whitchurch Brook and Glamorgan Canal.

## 5. Key findings

- The impact of aerial emissions upon levels of both  $^3\text{H}$  and  $^{14}\text{C}$  in sediments, invertebrates and fish at control sites just 10 km from the Amersham plc plant was below our detection limits. This indicates observable effects are restricted.
- The greatest activities of dissolved  $^3\text{H}$  were observed in small watercourses close to the Amersham plc plant (Glamorgan Canal and Whitchurch Brook), due to aerial emissions. These emissions had only a minor impact upon levels in the River Taff, due to a considerably greater dilution factor. The impact of liquid waste discharges, via the CSO, was minimal
- $^3\text{H}$  uptake by sediment, at sites solely affected by aerial deposition, was variable. Activities were low and only just above the detection limit on coarse grained sandy material from Whitchurch Brook, but consistently high on fine-grained silty organic rich mud from Glamorgan Canal. In the River Taff, upstream of the CSO, activities in the majority of sediment samples were below the limit of detection. Similar trends, albeit less marked, were discernable in the  $^{14}\text{C}$  data
- The impact of discharges from the CSO was clearly discernible in sediment samples collected downstream of the confluence with Whitchurch Brook, albeit that there was very large scatter in the total  $^3\text{H}$  and OBT data. It was not possible to identify any temporal trends

- $^3\text{H}$  assimilation in invertebrates was readily detectable at several sites solely affected by aerial deposition (levels in the order of  $10^2$  Bq/kg wet weight). CF values were in the range 4-27. Assimilation was, however, considerably greater at sites impacted by discharges from the CSO (average concentration elevated by  $\sim 10$  fold compared with that observed upstream of the CSO). To a first approximation, activities remained essentially constant throughout the study period. Levels of OBT at all sites were typically between one and two orders magnitude greater than those observed in the sediments.
- $^3\text{H}$  assimilation in fish was also readily detectable at several sites solely affected by aerial deposition. CF values were in the range 17-60. Levels in fish from Whitchurch Brook and Glamorgan canal were in the order of  $10^3$  Bq/kg wet weight.
- Data from the initial surveys in 2001 indicated extreme variability in  $^3\text{H}$  activities in bullheads obtained from sites downstream of the CSO in the River Taff. The 0+ age group ( $< 5$  cm) fish spawned in 2001 contained substantially lower activities than the largest ( $> 8$  cm) fish, likely to be more than two years old.
- The extreme variability, due to the presence of older fish spawned in or before 1999, was absent in subsequent surveys, presumably because bullheads are a short-lived species.
- The apparent impact of liquid wastes entering the River Taff, via the CSO, was markedly reduced in 2001 compared with that in 1999 and preceding years,

although the reason is not known. Its impact upon short-lived fish species such as bullheads is now minimal. Levels in some longer lived species remain elevated in older fish, most likely as a legacy of historically greater releases

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Hello David

Some notes for you.

I've marked comments and amends. Please discuss anything you're not happy with or where you have a better idea. **Accepted all of these**

For the figures, there is sometimes a lot of information, in both the graphs and the associated text. For a couple of examples, I've separated out the text in different ways so that the overall heading for each figure is more obvious. What do you think?  
**Agreed and changes made.**

The overall formatting is now rather messed up but there's no point fixing it yet.

A few more thoughts/comment below.

If the concentration in river water is so variable, we might be adding confusion to introduce the concept of CF. The activities aren't even enough to make the figures meaningful. **I feel we are throwing out the baby with the bathwater here. The concept is useful to provide an order of magnitude indication of the extent of bioassimilation. I have attempted to clearly identify the limits of its application.**

I've also wondered about the concept of sediment being washed away by bad weather/lots of water. The levels we're seeing downstream and in the undredged parts of the Bay are historic ie they've been there for a long time. The activity/sediment is therefore relatively stable. **My feeling is that the system is dynamic, as evidenced by the scatter in the data. I have revised the text to list the factors which may affect the survey data, rather than providing hypotheses.**

Between May 98 and April 2003 the nature of our discharges has remained constant. In April last year, most of the organic component was removed from the discharge. According to Welsh Water, there has been no change in the operation of the CSO in recent years. We can include these factual statements but I think we should remove speculative causes on other possibilities. Think the report should stick to presenting the data. **I have incorporated your suggestions into this draft**

Speak with you soon.

Julie

Verbal requests 31/8/04

- 1) Change name from GE Healthcare back to Amersham plc. **Done**
- 2) Take out a lot of the statistical information from the report (e.g. quoting average values for dissolved trit. in river water, calculating average values for OBT in invertebrates whilst excluding outliers). **Points accepted. See revised**

text regarding dissolved tritium and note that I have also no longer excluded outliers for the inverts.

- 3) Make description of models used for F-tests clear throughout
- 4) Julies partner (a statistician) feels that the data should be log transformed and replotted, particularly Fig. 7d. **Done**
- 5) Regression line for Fig. 9d “ $^{14}\text{C} = 0.15 * ^3\text{H} + 181$ ” should read “ $\log ^{14}\text{C} = 0.15 \pm ? * \log ^3\text{H} + 181 \pm ?$ ”. **Done**
- 6) Description of data in Figure 10c-check the statement “Age versus length indicated the data fitted well ( $r=0.064$ ) to a linear regression line of the form:  
  
Age =  $(0.36 \pm 0.03) * \text{length} - (3.5 \pm 0.1)$ ”. Should this read “ $r=0.964$ ” ? **Typo corrected**
- 6) Consider plotting Figure 11 as Log Scale..**Done**



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