WRC WATER RESEARCH CENTRE

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TRACE ELEMENTS IN WATER AND CARDIOVASCULAR DISEASE - POPULATION EXPOSURE TO METALS RELEASED FROM PIPING MATERIALS USED FOR WATER DISTRIBUTION IN THE UK (H0156C)

Final Report to the Department of the Environment 1977-1981

Project Leader: R F Lacey

Author: R J Bailey

January 1982

255-M

RESTRICTION: This report has the following restricted distribution:

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DOE CONTRACT REFERENCE: DGR/480/656

WRC ENVIRONMENT
MEDMENHAM LABORATORY: Henley Road,
Medmenham, P.O. Box 16, Marlow, Bucks SL7 2HD

TRACE ELEMENTS IN WATER AND CARDIOVASCULAR DISEASE - POPULATION EXPOSURE TO METALS RELEASED FROM PIPING MATERIALS USED FOR WATER DISTRIBUTION IN THE UK (HO156C)

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WRC ENVIRONMENTAL PROTECTION
Medmenham Laboratory, Henley Road,

SUMMARY

This is the final report on those parts of Project 0156 addressed to the following objectives:-

i) To investigate the relationship between the chemical composition (in particular the trace element content) of distributed waters at the tap and the characteristics of the source water and of the treatment, distribution and household plumbing systems.

ii) To investigate and evaluate methods of estimating population exposure to drinking water constituents, with particular reference to trace elements.

The concentrations of 26 elements in tap-water were surveyed at 40 houses in each of 25 towns, and have been combined with corresponding data on the consumption of tap-water based drinks to provide estimated intakes for middle-aged men. The concentrations of lead, copper and zinc in drinking water were also studied in relation to the quality of water from source, and pH appeared to be the most important water quality factor controlling levels of these metals in tap-water.

The ways in which lead and copper concentrations are related to stagnation time and flow-rate were investigated in detail at 7 locations. Stagnation time was shown to have an important effect on concentration but flow-rate less so.

A comparison of sampling methods for lead demonstrated the unreliability of discrete random samples for estimating mean intake. A proportional composite sampling device was evaluated on a limited scale.

A method of combining information on stagnation curves with information on the statistical distribution of inter-use stagnation times was developed and shown to provide a feasible approach to estimating the exposure of population groups.

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SUMMARY

1. INTRODUCTION

In 1977 the Department of the Environment (DOE) placed a 3 year contract with the Water Research Centre (WRC) for research into trace elements in drinking water, with special reference to the relationship between water hardness and cardiovascular disease. This contract was extended in 1980, when the modified objectives of the research became:-

i) To investigate the relationships between the chemical composition (in particular the trace element content) of distributed water at the tap and the characteristics of the source water and of the treatment, distribution and household plumbing systems.

ii) To investigate and evaluate methods of estimating population exposure to drinking water constituents with particular reference to trace elements.

iii) To investigate in collaboration with the MRC-funded team at the Royal Free Hospital, the relationships between the chemical composition of drinking water, the levels of cardiovascular risk factors measured in man and the rate of mortality from cardiovascular disease.

The medical aspects of this research (objective (iii)) have been the subject of separate reports(1,2) to the Department. The present report is the final report on the work undertaken towards objectives (i) and (ii) in the four years up to March 1981.

Objectives (i) and (ii) were pursued in four separate but related investigations. The first aimed at investigating the occurrence of a wide range of trace elements in drinking water sampled at houses in 25 British towns. Information was also obtained about the drinking habits of the occupants of the houses. The remaining investigations were more detailed but narrower in scope and smaller in scale. They concentrated on just two of the more important health related trace metals associated with plumbing systems - lead and copper. The second and third investigations attempted to quantify the variability of lead and copper in drinking water drawn from consumers' taps. The fourth study developed a method of modelling the variations in tap water lead levels from which indirect estimates of population exposure can be derived.

Appendices 1 to 4 give detailed descriptions of the work undertaken in the four project areas together with the results and conclusions drawn from each separate investigation. This report describes the main features of the programme as a whole.

2. WORK UNDERTAKEN

2.1. WATER SAMPLING IN THE BRITISH REGIONAL HEART STUDY

The British Regional Heart Study is a major epidemiological study of cardiovascular disease in the UK, being undertaken by the Royal Free Hospital School of Medicine, London. The study includes a cross-sectional survey of middle-aged men, some 300 individuals in each of 25 towns. WRC has undertaken tap-water sampling for about 40 of these men in each town. At each house three samples were taken: 'first draw' (after water had stood in the pipes overnight), 'random daytime' and 'fully flushed'. These samples were each analysed for the 26 elements listed below:-

Aluminium
Barium
Beryllium
Bismuth
Boron
Cadmium
Calcium
Chromium
Cobalt
Copper
Iron
Lead
Lithium
Magnesium
Manganese
Molybdenum
Nickel
Potassium
Silicon
Silver
Sodium
Strontium
Titanium
Vanadium
Zinc
Zirconium

At the same time as the medical survey, the men selected for water sampling were each taken through a questionnaire about their consumption of tap-water based drinks and habits of water use. The questionnaire was designed to reveal for each man:-

i) The total daily intake of tap-water.

ii) The proportion of tap-water drunk as tea, coffee, plain water and other drinks.

iii) The proportion of tap-water drunk in each of the 3 sampling categories: 'first draw', 'random daytime' and 'fully flushed'.

iv) Whether water was drawn for drinking purposes from taps other than the kitchen cold tap.

2.2. AN INVESTIGATION OF THE FACTORS AFFECTING LEAD AND COPPER LEVELS IN DRINKING WATER

The effects of stagnation time and flow rate on the concentrations of lead and copper in water drawn from pipes made of those materials were investigated for seven pipes (3 lead, 4 copper) in different parts of the country. Experiments were designed to answer the following specific questions for lead, and similar questions for copper.

(i) How does flow-rate affect the concentration of lead in tap-water when a lead service pipe is continuously flushed to reach steady state conditions?

(ii) How does the stagnation time of water in the service pipe affect the concentration of lead in tap-water? Can the relationship between stagnation time and lead concentration be described by a simple mathematical model?

(iii) Does the flow-rate at which stagnation samples are taken have any effect on the lead concentration?

(iv) Does the proportion of insoluble lead material in tap-water vary with flow-rate?

2.3. SAMPLING VARIATION AND PROPORTIONAL SAMPLING METHODS

The within-house variability of lead concentrations in different types of water sample has been studied for a set of 16 random daytime and 16 first draw samples taken from each of 11 houses in Glasgow.

One method of overcoming the problems of variability of individual samples is to use a proportional composite sampling device attached to the consumer's tap. WRC has experimented with the type of proportional sampler designed by RID† in the Netherlands. These samplers were installed in eight co-operative households over the period of a week to check whether the households would accept the sampler and use it correctly, and to compare estimates of exposure derived from the proportional sampler with estimates derived from discrete sampling and manual recording of water use.

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† Rijksinstituut voor Drinkwatervoorziening (National Institute for Drinking Water Supply), Leidschendam, The Hague, Netherlands.

2.4. THE RELATIONSHIP BETWEEN TAP-WATER LEAD LEVELS AND WATER USE PATTERNS

Stagnation curves for lead in tap-water, which were established from the work described in Section 2.2, were combined with a frequency distribution of stagnation times to derive the frequency distribution of lead concentrations in drinking water. Summary statistics, such as the mean or median lead concentrations in drinking water, could be obtained from this distribution. For input data we took a frequency distribution of overnight stagnation times available from the water sampling survey associated with the British Regional Heart Study (Section 2.1). In the absence of information on stagnation times throughout the day, an informal survey was carried out by placing diaries with the households of several members of WRC staff.

3. RESULTS AND DISCUSSION

3.1. A SURVEY OF TRACE ELEMENT LEVELS IN DRINKING WATER IN 25 BRITISH TOWNS (APPENDIX 1)

Mean levels for the first draw and fully flushed concentrations of 26 trace elements estimated for aggregates of 12 soft water towns and 11 hard water towns are shown in Table 1. More detailed town-by-town statistical summaries are provided in Appendix 1 for the first draw, random daytime and fully flushed concentration levels of five of the more important trace metals associated with distribution systems, namely lead, copper, zinc, iron and manganese.

The relative quantities of tap-water and different beverages consumed at home by the sub-sample of Regional Heart Study respondents were as follows:-

Tea	68%
Coffee	20%
Tap-water	10%
Other drinks	2%

The total daily volume of tap-water consumed in all the categories listed above was 1.25 litres per head.

The relative volumes of tap-water drunk at home in the three sampling categories were:-

Table 1 Average trace element concentrations in first-draw (FD) and fully flushed (FF) water and exposures for middle-aged men

Element	Soft water towns			Hard water towns		
	FD	FF	Intake	FD	FF	Intake
	µg/l	µg/l	µg/day	µg/l	µg/l	µg/day
Ag	3 b	3 b	-	3 b	3 b	-
Al	190	180	230	26 b	27 b	-
B	11	9	13	59	59	74
Ba	45	43	55	88	85	110
Be	0.1 b	0.1 b	-	0.1 b	0.1 b	-
Bi	41 b	48 b	-	42 b	51 b	-
Ca	18,000	17,000	22,000	92,000	90,000	110,000
Cd	2 b	2 b	-	2 b	2 b	-
Co	5 b	5 b	-	5 b	5 b	-
Cr	3 b	3 b	-	5	4	5
Cu	140	24	77	300	14	110
Fe	110	100	130	40	31	44
K	1,000	970	1,200	3,300	3,200	4,100
Li	3 b	3 b	-	7	7	9
Mg	3,300	3,300	4,100	12,000	12,000	15,000
Mn	20	22	27	4	5	6
Mo	37 b	37 b	-	45 b	54 b	-
Na	8,300	8,100	10,000	28,000	28,000	35,000
Ni	20 b	18 b	-	18 b	16 b	-
Pb	47	11	24	14	3	9
Si	2,000	2,000	2,500	4,600	4,500	5,700
Sr	57	56	70	350	340	420
Ti	4	4	5	16	14	18
V	4	4	5	12	12	14
Zn	63	19	39	110	9	42
Zr	4 b	4 b	-	5 b	5 b	-

b means below the analytical limit of detection

First draw	6%
Random daytime	57%
Fully flushed	37%

95% of the total volume of water drunk at home was drawn from the kitchen cold tap; the rest being taken from hot taps or other cold taps.

Information on the average volume of water consumed was combined with information on the mean levels of trace elements in tap-water to give the crude estimates of daily intakes of trace elements shown in the third and sixth columns of Table 1.

The data from the trace metal survey revealed which elements were likely to be associated with service pipe and plumbing materials. The comparison of first draw and fully flushed samples confirmed that large increases of between 300 and 2000% in lead and copper levels occur during overnight stagnation in the presence of pipes of these materials. Zinc levels increase by about 1200% in hard water areas, but only by about 230% in soft water areas. Cadmium levels in soft water towns increase by about 40% in first draw samples compared with a 17% increase in soft water towns. However, the cadmium comparison was made for only those samples in which cadmium was detected; about 5.5% of the total number analysed. Iron, vanadium and zirconium show statistically significant increases of between 7 and 14% in first draw samples in soft water areas. Beryllium, bismuth, manganese and molybdenum show significant decreases in first draw samples. Beryllium and manganese show statistically significant decreases of 19% and 10% respectively, only in soft water areas; molybdenum shows a 22% decrease in hard water towns and bismuth a 45% decrease in both hard and soft water towns. The remaining trace metals: silver, aluminium, cobalt, chromium, nickel and titanium do not show statistically significant changes in concentration after prolonged contact with pipes.

A multiple regression analysis, using a relationship of the general form:-

$$\log_e [\text{metal}] = \alpha + \beta\text{pH} + \sum_i \gamma_i \log_e x_i + \varepsilon$$

was carried out to discover which of the major water quality parameters (x_i) most influenced the levels of lead, copper and zinc in tap-water. This revealed pH as probably the most important factor for all three metals. In the case of lead, however, pH only appeared to exert a significant influence in soft water towns.

3.2. FACTORS AFFECTING LEAD AND COPPER LEVELS IN DRINKING WATER (APPENDIX 2A AND 2B)

The results of the stagnation experiments showed a very marked dependence of both lead and copper on stagnation time. Concentrations increased rapidly over the first few hours and levelled off somewhere between 10 and 20 hours at concentrations up to 20 times that of the original flushing base level concentration. A simple mathematical function of the log-hyperbolic form can be used to describe the stagnation curves obtained experimentally.

The dependence of lead and copper levels on flow-rate under steady state continuous flow conditions was less strong. It was found that metal levels usually decreased with increasing flow-rate in the range 0.5 to 3 litres $\min^{-1}$ and then levelled off. In some cases lead, but not copper, levels showed a tendency to rise again at flow-rates above about 7 litres $\min^{-1}$. This was attributed to scouring of particulate material from the pipe walls under very turbulent flow conditions.

3.3. SAMPLING VARIATION AND PROPORTIONAL SAMPLING METHODS (APPENDIX 3)

From the data obtained at 11 houses in Glasgow the within-house variation of random daytime samples appeared to be approximately log-normally distributed; the standard deviation of the logged (base 10) results being about 0.21. The distribution of repeat first draw samples approximated the log-normal distribution less well than did the distribution of random daytime samples but the within-house variation was less; the standard deviation of the logged (base 10) results being about 0.15.

The results from this work imply that a single random daytime sample is capable of estimating the mean daytime lead concentration at a house only to within a factor of about 3. A single first draw sample will give a slightly more reliable estimate of the mean first draw lead concentration but this parameter will overestimate exposure levels as first draw water is rarely drunk. (Section 3.1). It is clear from these results that taking either a single random daytime or a single first draw sample from a house cannot be relied on to give an accurate measure of exposure.

The practical assessment of the proportional sampling device originally developed by RID in the Netherlands revealed that, although consumers accepted the presence of the sampler on their kitchen taps in spite of the inconvenience caused, they did not always use it properly. This was deduced from the observation that the volume of water consumed (estimated from a diary kept by the house occupants) was usually less than the volume of water in the sample bottle. Since this suggests that water was sampled on occasions of water use other than for drinking, errors could result in estimates of exposure derived. No clear result emerged from the experiment designed to compare the exposure values obtained from the proportional sampler with those calculated from drinking habits and discrete sample analyses. There were indications that lead exposure levels estimated from the sampler were higher than those predicted from discrete samples. However, the size of this investigation was too small to be conclusive.

It is recommended that future work on proportional sampling methods should concentrate on designing a sampler that minimises the possibilities of human error and which is also more acceptable to the consumer.

3.4. RELATING TAP-WATER LEAD TO WATER USE PATTERNS (APPENDIX 4)

We have seen from Section 3.3 that direct assessment of exposure using discrete tap-water samples, such as the random daytime sample, was found to be unreliable because of the large within-house variation in levels of trace metals in tap-water. Proportional sampling devices, which produce a single composite sample representative of the mean metal concentration weighted by the volume of water drawn for consumption, are likely to give more reliable individual exposure estimates. However, they are expensive to install and can be misused by the consumers.

Appendix 4 shows how the problem of variation in levels of trace metals in tap water can be approached by separating its two main components:-

i) The variation in metal levels with contact time in the pipes, which was investigated in Appendix 2.

ii) The variation in the contact times themselves resulting from the consumers' water use patterns.

Levels of exposure to trace metals can then be predicted, on a population basis rather than an individual basis, by combining information on stagnation curves with that for typical consumers' patterns of water use. This indirect method therefore provides an alternative to direct assessment of population exposure by aggregating individual exposures estimated from discrete samples. In addition, it offers scope for increasing the accuracy of future estimates of population exposure as improved information on household water use patterns and theoretical methods of predicting stagnation curves* become available.

The approach also leads to the concept of fixed stagnation time sampling methods which would be far more informative for the same amount of sampling effort than the random sampling methods employing discrete samples, which are currently used in the UK.

4. OVERALL CONCLUSIONS

i) Information has been gathered about the concentrations of trace metals in water sampled at taps in 25 towns in Britain. This has been combined with volume data to give average estimates of exposure for middle-aged men on a town by town basis.

ii) Estimation of exposure to plumbing metals is difficult because of the large temporal variability in concentrations, arising mainly from

a) the stagnation performance of the pipe system,

b) the water use patterns of the consumer.

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*Recent work by Wagner and Kuch at the Engler-Bunte Institute for Water Chemistry, University of Karlsruhe, West Germany has demonstrated the feasibility of using mass transfer theory to predict stagnation curves for lead in drinking water.

iii) Discrete water samples are therefore unreliable for estimating the exposure of a particular individual. The effects of (a) and (b) are better combined physically by a proportional composite sampling device. The acceptability of proportional samplers to consumers has not however been tested on a large enough scale in the UK to make a confident recommendation.

iv) For estimating the exposure of population groups there is the alternative approach of combining separately acquired information about the relevant populations of (a) and (b). This is theoretically feasible and attractive in practice, if the degree of approximation can be tolerated in the context in which the results are to be used.

REFERENCES

1. POWELL, P., PACKHAM, R.F., LACEY, R.F., RUSSELL, P.F., SHAPER, A.G., POCOCK, S.J., and COOK, A.G. Water Quality and Cardiovascular Disease in British towns. WRC Technical Report. (In preparation).

2. LACEY, R.F. Changes in Water Hardness and Cardiovascular Death-Rates. WRC Technical Report TR 171, 1981.

APPENDIX 1

A SURVEY OF TRACE ELEMENT LEVELS IN DRINKING WATER IN 25 BRITISH TOWNS

CONTENTS

APPENDIX 1

A SURVEY OF TRACE ELEMENT LEVELS IN DRINKING WATER IN 25 BRITISH TOWNS

1. INTRODUCTION

This Appendix describes the survey of trace element levels in drinking water which was carried out in conjunction with the British Regional Heart Study (RHS), an epidemiological study of middle-aged men in 25 towns in England, Scotland and Wales.

The aims of the survey were:-

- To investigate trace element levels in tap-water in towns covering a wide range of water qualities.
- To estimate the exposure of the RHS subjects to trace elements in their drinking water.
- To determine which trace elements are either leached out or deposited on contact with service pipes or plumbing.
- To determine which are the most important water quality parameters affecting the levels of certain plumbing related trace metals in tap-water.

In Great Britain there are marked regional variations in the death-rate from cardiovascular disease (CVD). The highest rates are found in Scotland, the North of England and Wales; the lowest rates in the South and South-East England. The aim of the Regional Heart Study, which is being run by the Royal Free Hospital's Department of Clinical Epidemiology and General Practice, is to determine whether these regional differences are associated with any of the established risk factors for cardiovascular disease, for example, hypertension, smoking, raised blood lipid levels etc, or with environmental factors such as climate or water hardness.

Epidemiological studies in Great Britain, based on analysis of mortality data around the times of the 1951, 1961 and 1971 censuses, have shown a statistically significant association between water hardness and death-rate from CVD: the highest rates in soft water towns and the lowest rates in hard water towns.

2. DESIGN

2.1. MEDICAL SURVEY

Twenty-five towns were selected in England, Scotland and Wales to represent the range of CVD mortality rates and water hardness and also to cover all regions of the country. The map of Great Britain (Figure 1) shows the geographical distribution of these 25 towns. Other criteria for choosing the towns were:-

i) their populations should be between 40,000 and 120,000,
ii) they should be separate entities, away from major conurbations.

In each town, about 300 men aged between 40 and 59 years were selected at random for medical examination from the age/sex register of a large, representative general practice.

The results described in this report are confined to the WRC water quality survey. The medical aspects of the Regional Heart Study are outside the scope of this contract. However, a description of the design of the medical survey, and the results so far obtained, is given in a recent paper by Shaper and others in the British Medical Journal(1).

2.2. WATER SAMPLING SURVEY

The Water Research Centre undertook a survey of drinking water quality, run in conjunction with the medical survey. At each town visited during the study a sub-sample of 40 men was selected from the 300 called for the medical examination. Each man in the sub-sample was taken through a questionnaire on his drinking water consumption habits. This questionnaire was designed to reveal the following information for each man:-

1) The total daily intake of tap-water.
2) The proportion of tap-water drunk in each of 4 beverage categories, i.e., tea, coffee, plain water and other tap-water-based drinks.
3) The proportion of tap-water drunk in each of 3 sampling categories, i.e., first draw, random daytime, fully flushed.
4) Whether any water was drawn for dinking purposes from taps other than the cold tap in the kitchen.

The subject was then given a bottle to take home with him and instructed to fill it from the kitchen cold tap first thing the next morning before using

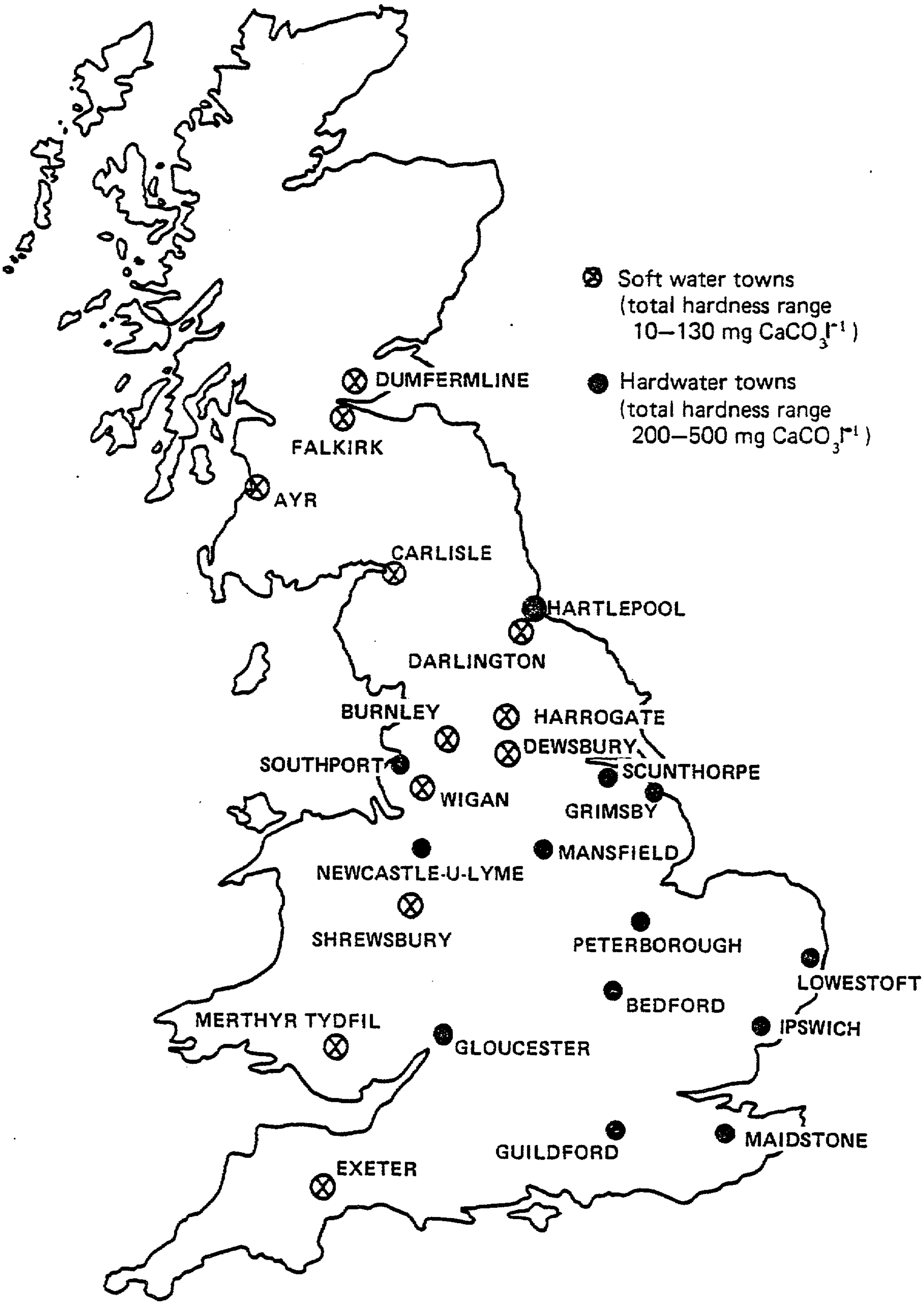

Fig. 1. Regional heart study towns

water for any other purpose anywhere in the house. This was then the 'first draw' tap-water sample. WRC samplers visited each house the next day to collect this sample and take 3 others as follows:-

i) A random daytime sample, which was taken without any preflushing of the tap.

ii) A fully flushed sample, taken after flushing at least 5 service pipe volumes of water.

iii) A non-acidified sample for determination of hardness and other major water quality parameters.

Additionally, the following information and measurements were obtained at each house:-

i) Overnight stagnation time, recorded by the consumer on taking the the first draw sample.

ii) Stagnation time before taking the random daytime sample - a subjective estimate based on the consumer's recall.

iii) The length and type of material of the communication pipe (water undertaking owned), supply pipe (consumer owned) and the internal plumbing.

iv) The outlet last used and purpose of last use.

v) Whether the kitchen cold tap was supplied from a storage tank (a rare occurrence).

vi) The type of hot water system.

vii) Whether the kitchen cold tap was a mixer tap, plastic tap or was fitted with a spray or swirler nozzle.

viii) The number of separate dwellings sharing a service pipe (shared services are common in inner urban areas).

ix) Age and type of property.

x) Measurements of pH, conductivity and water-temperature were taken during the visit to each house.

3. METHODS

3.1. SAMPLING

First draw, random daytime and fully flushed samples were collected in 1 litre acid washed polythene bottles and acidified to pH 1 with nitric acid of 'Aristar' purity.

Samples for the determination of hardness and major water quality parameters were collected in 1 litre, acid washed polythene bottles after rinsing twice with the sample water. These samples were not acidified.

pH was measured using a portable EIL 3050 pH meter.

Conductivity was measured using a portable Simac 63 conductivity meter.

Temperature was measured using an ordinary mercury-in-glass thermometer held in the stream of water <u>after</u> prolonged flushing. The temperature measured, therefore, represented that of the water in the mains.

Service pipe lengths were measured by pacing the line assumed to be taken by the pipe. At all houses the assistance of a Water Authority Inspector was available and his local knowledge of plumbing installations proved very useful in obtaining this and other information.

3.2. CHEMICAL ANALYSIS

Samples were analysed for 26 trace elements simultaneously using an inductively coupled plasma spectrometer type ARL 137. The performance of this instrument has been reported recently(2).

Lead was determined separately because the plasma spectrometric technique did not satisfy the detection limit required for this metal. The method used was atomic absorption spectrometry and the performance characteristics of the instrument are the same as those described in Appendix 2A (Section 2.3) except that the detection limit was improved from 5 μg Pb l^{-1} to 2 μg Pb l^{-1}.

Major water quality parameters were measured using standard auto-analyser techniques or plasma spectrometry depending on which was easier, provided it gave satisfactory results. A list of these parameters is given below showing which were measured by plasma spectrometry:-

Total hardness*
Alkalinity
Nitrate/Nitrite
Silica (SIO_2, molybdate reactive)
Chloride
Sulphate

Silicon (Si) (plasma)
Calcium (plasma)
Magnesium (plasma)
Sodium (plasma)
Potassium (plasma)

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* For the second half of the study, the standard method for total hardness was replaced by the plasma method in which the hardness was calculated from the sum of the calcium and magnesium values.

3.3. STATISTICAL ANALYSIS

The statistical analysis of the water quality data falls into five separate areas:-

i) Analysis of the results of the questionnaire on drinking water consumption habits.

ii) A town-by-town statistical summary of all important trace elements and other water quality parameters.

iii) An estimate of the mean daily intake of important trace elements on a town-by-town basis for typical middle-aged men. This estimate is obtained by combining information on mean levels of trace elements with information on water consumption.

iv) A paired comparison of first-draw and fully-flushed samples to reveal the extent to which metals or other elements are either leached from or deposited in the service pipe after a prolonged contact time.

v) A multiple regression analysis of the data by source to reveal which are the most important major water quality parameters controlling the levels of trace metals in drinking water.

4. RESULTS

4.1. DRINKING WATER CONSUMPTION HABITS

Table 1 summarises the drinking habits of the sub-sample of middle-aged men taking part in the water sampling survey. These results were obtained from the combined data for the 25 towns, with one town (Hartlepool) repeated making a total of 26 separate town units of data. Table 1A shows the relative quantities of tap-water and different beverages consumed. Table 1B shows the relative amounts of tap-water drunk in the 3 different sampling categories. Table 1C shows how water consumed is distributed between the different outlets in the house. All three tables give a breakdown of the mean total daily tap-water intake for middle-aged men, which is 1.25 litres lead^{-1} day^{-1}: 0.91 litres drunk at home, and 0.34 litres drunk elsewhere. It should be noted that the relative proportions of tap-water drunk in the 3 sample categories (Table 1B) differ slightly from those given in the Summary Report because the results are now based on data from all 25 towns; data were only available from 18 towns for the Summary Report.

4.2. SUMMARY OF WATER QUALITY DATA

4.2.1. Major water quality parameters

Table 2 shows the 23 towns for which data are currently available, the

Table 1A. Quantities of tap water and beverages consumed.

Breakdown	Quantity(1/head/day)	Standard error	95% conf.limits	% of total
Tea	0.85	0.019	0.82-0.89	68.3
Coffee	0.25	0.010	0.23-0.27	20.0
Tap water	0.12	0.010	0.10-0.14	9.5
Other	0.03	0.003	0.02-0.04	2.2
TOTAL	1.25	0.019	1.21-1.29	100.0

Table 1B. Relative quantities of tap water drunk in 3 sample categories.

Breakdown	Quantity(1/head/day)	Standard error	95% conf.limits	% of total
Home:- First draw	0.06	0.004	0.05-0.06	4.5 (6% of home total)
Random day	0.54	0.018	0.50-0.60	43.0 (57% of home total)
Fully flushed	0.35	0.016	0.32-0.39	28.4 (37% of home total)
HOME TOTAL	0.95	0.017	0.92-0.98	75.9
Elsewhere	0.30	0.008	0.28-0.32	24.1
OVERALL TOTAL	1.25	0.019	1.21-1.29	100.0

Table 1C. Relative quantities of tap water drunk at different outlets in the house.

Breakdown	Quantity(1/head/day)	Standard error	95% conf.limits	% of total
Kitchen tap	0.90	0.017	0.87-0.93	72.0 (94.7% of home total)
Other cold tap	0.01	0.001	0.00-0.01	0.5 (0.7% of home total)
Hot tap	0.04	0.007	0.00-0.01	3.4 (4.6% of home total)
HOME TOTAL	0.95	0.017	0.92-0.98	75.9
Elsewhere	0.30	0.008	0.28-0.32	24.1
OVERALL TOTAL	1.25	0.019	1.22-1.29	100.0

Table 2 - Date of sampling and mean values of the major water quality parameters

	Sampling date	Total hardness mg CaCO3/l	Alkalinity mg CaCO3/l	pH	Calcium mg/l	Magnesium mg/l	Sodium mg/l	Potassium mg/l	Chloride mg/l	Sulphate mg/l	Nitrate mg N/l
Ayr	Jul 1979	12.7	5.0	6.2	3.6	0.9	3.8	0.3	7.1	6.8	0.1
Burnley	Nov 1978	39.5	21.2	8.0	9.9	2.7	11.8	0.8	9.2	20.8	0.5
Carlisle	Nov 1979	124.0	67.5	7.4	42.1	5.0	5.2	0.9	8.2	61.7	1.0
Darlington	Oct 1979	89.8	61.1	7.8	30.3	3.5	4.8	1.0	9.1	43.3	0.5
Dewsbury	Mar 1979	39.1	12.8	9.3	10.1	3.3	16.7	1.1	29.0	22.9	1.0
Dunfermline	Sep 1979	45.2	48.8	8.1	13.0	2.9	4.4	0.4	4.8	18.0	0.3
Exeter	Feb 1979	72.7	25.1	----	20.9	4.8	13.8	2.2	29.1	24.5	3.8
Falkirk	Apr 1979	31.8	8.2	---	9.8	1.7	5.0	0.6	10.2	13.8	0.6
Harrogate	Feb 1978	64.2	33.1	8.3	17.2	5.2	9.1	1.3 /	15.1	28.1	0.8
Merthyr Tydfil	Jul 1978	33.7	17.8	7.3	12.4	1.3	4.3	0.5	8.3	10.1	0.2
Shrewsbury	Mar 1978	103.0	41.3	7.9	30.9	5.6	10.4	2.1	23.7	42.9	2.7
Wigan	Apr 1980	28.3	19.1	7.5	7.7	2.0	7.6	0.6	8.6	10.9	0.6
Bedford	Mar 1980	302.1	193.1	7.6	106.1	8.6	38.0	6.0	46.8	156.8	9.2
Gloucester	Jun 1979	237.3	130.9	8.0	67.4	15.9	31.5	4.2	56.7	76.4	5.8
Grimsby	Feb 1980	268.7	209.6	7.5	95.9	8.1	13.3	2.0	22.7	39.3	5.6
Guildford	Oct 1978	263.2	190.3	7.7	91.0	3.3	15.6	2.7	27.4	44.0	7.5
Ipswich	May 1979	382.6	278.6	---	134.4	11.5	40.9	4.8	80.6	80.7	6.5
Maidstone	Dec 1979	293.6	236.2	7.3	109.5	4.3	18.1	2.5	32.4	37.2	3.5
Mansfield	May 1978	220.9	82.7	7.5	44.0	27.6	30.4	2.8	76.0	71.2	7.7
Newcastle-u-Lyme	Dec 1978	208.8	165.2	8.2	57.4	15.4	10.0	2.8	18.0	26.7	4.4
Peterborough	Dec 1977	320.0	256.0	7.1	134.3	9.9	15.1	2.5	38.9	104.0	0.2
Scunthorpe	May 1980	289.1	211.2	7.1	105.7	6.9	68.9	2.6	50.1	123.0	11.7
Southport	Jun 1978	226.2	144.2	7.3	48.7	23.3	21.7	2.5	33.7	80.7	0.3

dates of sampling and the average values of 10 major water quality parameters. The towns fall naturally into two classes based on total hardness. In Table 2 the 12 soft water towns (Ayr to Wigan) are listed first followed by the 11 hard water towns (Bedford to Southport). The range of mean total hardness for the soft water towns is 13 to 124 mg $CaCO_3$ l^{-1}; for the hard water towns it is 209 to 383 mg $CaCO_3$ l^{-1}. Thus there is a clear division between the two groups of towns of 85 mg $CaCO_3$ l^{-1} total hardness.

4.2.2. Missing data

Results are reported for only 23 of the 25 towns sampled. Data for Hartlepool and Lowestoft (mean total hardness 500 and 380 mg $CaCO_3$ l^{-1} respectively) are not included in any of the statistical analyses because of problems with the plasma emission spectrographic analysis when applied to these very hard waters. Although the analyses are being repeated, the results were not available in time for inclusion in this report.

pH values for Exeter, Falkirk and Ipswich are missing from Table 2. The reason for this was breakdown of the pH electrode during sampling.

4.2.3. Trace elements

The following elements were determined by plasma emission spectrometry:-

Element	Detection limit ($\mu g l^{-1}$)
Silver	6
Aluminium	36
Boron	2
Barium	6
Beryllium	0.2
Bismuth	78
Calcium	34
Cadmium	4
Cobalt	10
Chromium	4
Copper	4
Iron	4
Potassium	70
Lithium	4
Magnesium	2
Manganese	2
Molybdenum	58
Sodium	70
Nickel	24
Silicon	26
Strontium	> 2
Titanium	4
Vanadium	4
Zinc	6
Zirconium	8

Lead was determined exclusively by atomic absorption spectrometry. The detection limit was 2 $\mu g l^{-1}$.

The grand means for these 26 elements over the 2 aggregates of 12 soft water towns and 11 hard water towns are shown in Table 3. Exposure estimates, in terms of mean daily intakes, are also given for all the elements in Table 3.

Five trace metals were considered to be of special importance in this study because they were either health related or known to cause taste and corrosion problems by leaching from mains or plumbing materials. These metals were:

Lead
Copper
Zinc
Iron
Manganese

Therefore summaries, on a town-by-town basis, for all concentrations of these elements are given in Tables 4 to 8. Results for all three sample categories have been included in these Tables (In Table 3 the random daytime results were omitted for conciseness). It should be noted that for lead, copper and zinc the mean random daytime concentration value usually lies somewhere between the first-draw and fully-flushed values. It is important to note that for lead or copper the estimates of the means apply to only those houses with lead or copper pipe present in the service connection or household plumbing. It has been found from previous studies at the WRC and confirmed in this study, that levels of lead or copper are negligible in the absence of pipes of the corresponding metal. An important exception to this is the presence of lead-based solders in copper pipe joints(3,4,5). However, problems are usually only encountered in large plumbing installations and no attempt was made to assess the effect of soldered joints in this study. Table 9 summarises, for each town, the number of houses in which the four common pipe materials - lead, copper, polythene and galvanised iron - were present in some part of the service connection or household plumbing.

4.3. TRACE ELEMENT INTAKES (EXPOSURE ESTIMATES)

From the water consumption questionnaire, the division of the daily volume of water drunk at home into the three sampling categories: first-draw, random daytime and fully-flushed, was 6% 57% and 37% respectively.

The mean daily intakes of the five metals: lead, copper, zinc, iron and

Table 3 Average trace element concentrations in first-draw (FD) and fully flushed (FF) water and exposures for middle-aged men

Element	Soft water towns			Hard water towns		
	FD µg/l	FF µg/l	Intake µg/day	FD µg/l	FF µg/l	Intake µg/day
Ag	3 b	3 b	-	3 b	3 b	-
Al	190	180	230	26 b	27 b	-
B	11	9	13	59	59	74
Ba	45	43	55	88	85	110
Be	0.1 b	0.1 b	-	0.1 b	0.1 b	-
Bi	41 b	48 b	-	42 b	51 b	-
Ca	18,000	17,000	22,000	92,000	90,000	110,000
Cd	2 b	2 b	-	2 b	2 b	-
Co	5 b	5 b	-	5 b	5 b	-
Cr	3 b	3 b	-	5	4	5
Cu	140	24	77	300	14	110
Fe	110	100	130	40	31	44
K	1,000	970	1,200	3,300	3,200	4,100
Li	3 b	3 b	-	7	7	9
Mg	3,300	3,300	4,100	12,000	12,000	15,000
Mn	20	22	27	4	5	6
Mo	37 b	37 b	-	45 b	54 b	-
Na	8,300	8,100	10,000	28,000	28,000	35,000
Ni	20 b	18 b	-	18 b	16 b	-
Pb	47	11	24	14	3	9
Si	2,000	2,000	2,500	4,600	4,500	5,700
Sr	57	56	70	350	340	420
Ti	4	4	5	16	14	18
V	4	4	5	12	12	14
Zn	63	19	39	110	9	42
Zr	4 b	4 b	-	5 b	5 b	-

b means below the analytical limit of detection

Table 4 - LEAD (ug/l)

	First draw			Random daytime			Fully flushed			Intake (ug/day)
	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	mean
Ayr	187.7	559.1	930.4	85.0	305.1	525.1	70.7	147.1	223.5	327.3
Burnley	16.5	24.2	31.8	6.5	11.8	17.2	2.4	4.4	6.4	12.3
Carlisle	21.0	47.9	74.7	7.1	27.4	47.8	2.0	7.2	12.4	26.5
Darlington	11.3	30.9	50.4	<2.0	22.0	45.3	4.8	8.1	11.4	21.8
Dewsbury	<2.0	65.5	158.1	<2.0	8.3	19.9	<2.0	<2.0	<2.0	11.4
Dunfermline	13.9	27.8	41.7	7.9	20.0	32.1	4.8	8.6	12.4	20.3
Exeter	19.0	30.2	41.5	10.3	18.2	26.1	<2.0	9.5	17.9	19.6
Falkirk	4.1	9.1	14.1	<2.0	4.1	7.8	<2.0	<2.0	2.5	4.4
Harrogate	35.1	57.8	80.5	15.3	24.8	34.3	4.8	6.6	8.5	25.1
Merthyr Tydfil	8.2	48.1	88.1	7.8	27.6	47.3	3.9	10.5	17.2	28.1
Shrewsbury	2.5	9.4	16.4	<2.0	3.7	7.6	<2.0	<2.0	<2.0	4.0
Wigan	83.7	143.6	203.5	40.6	70.1	99.7	20.3	32.7	45.2	75.9
Bedford	9.9	27.2	44.6	5.7	11.7	17.7	2.6	5.0	7.4	12.7
Gloucester	27.1	41.8	56.5	15.8	31.1	46.4	6.7	12.2	17.7	31.0
Grimsby	6.4	11.1	15.8	3.7	8.4	13.1	<2.0	2.6	3.6	8.0
Guildford	<2.0	5.9	14.2	<2.0	3.0	6.8	<2.0	<2.0	2.6	3.3
Ipswich	8.3	22.3	36.3	6.5	15.2	24.0	<2.0	5.6	10.4	15.1
Maidstone	---	---	---	---	---	---	---	---	---	---
Mansfield	4.4	12.1	19.9	<2.0	5.6	9.4	<2.0	<2.0	2.7	5.7
Newcastle-under-Lyme	18.1	25.6	33.0	10.4	19.9	29.3	3.8	4.9	6.1	18.3
Peterborough	<2.0	48.5	488.7	<2.0	35.5	511.7	<2.0	7.5	22.4	33.3
Scunthorpe	14.8	61.2	107.6	12.7	31.0	49.3	4.6	8.9	13.2	30.8
Southport	16.6	23.2	29.9	11.8	17.2	22.5	3.2	4.7	6.1	16.1

Table 5 - COPPER (ug/l)

	First draw			Random daytime			Fully flushed			Intake (ug/day)
	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	mean
Ayr	650.1	827.3	1004.6	353.9	485.3	616.6	70.5	96.5	122.4	452.4
Burnley	20.9	30.5	40.1	8.0	13.2	18.3	<4.0	<4.0	<4.0	13.1
Carlisle	15.8	81.6	147.4	4.9	26.3	47.7	<4.0	<4.0	5.4	26.7
Darlington	21.2	37.6	54.0	32.6	45.7	58.7	64.9	88.0	111.1	76.1
Dewsbury	7.7	14.0	20.3	4.5	5.6	6.8	<4.0	<4.0	<4.0	6.3
Dunfermline	80.6	102.5	124.5	83.8	104.9	126.1	23.0	34.1	45.1	98.2
Exeter	32.7	53.4	74.1	9.1	22.7	36.3	<4.0	4.1	5.8	22.0
Falkirk	17.3	31.4	45.4	5.8	10.6	15.3	<4.0	<4.0	4.5	11.5
Harrogate	38.7	71.4	104.0	17.0	28.6	40.2	5.5	7.3	9.2	29.1
Merthyr Tydfil	154.3	266.4	378.5	83.9	136.6	189.2	16.9	31.5	46.1	131.9
Shrewsbury	20.5	32.1	43.7	8.9	16.0	23.1	<4.0	4.4	6.6	15.8
Wigan	97.3	169.8	242.3	40.0	80.1	120.1	7.1	11.7	16.4	75.2
Bedford	282.3	396.3	510.3	125.4	198.6	271.8	4.8	7.0	9.2	174.5
Gloucester	68.6	126.4	184.2	28.5	54.3	80.0	6.8	10.2	13.7	52.9
Grimsby	76.7	106.5	136.3	16.1	31.3	46.4	<4.0	<4.0	4.2	31.8
Guildford	216.7	306.7	396.7	83.6	113.4	143.2	14.4	20.6	26.7	113.3
Ipswich	375.9	579.1	782.3	154.8	263.0	371.2	19.4	29.5	39.7	244.5
Maidstone	60.5	251.2	442.0	43.7	71.7	99.7	<4.0	5.2	7.6	72.3
Mansfield	221.4	288.5	355.5	85.4	129.2	172.9	19.2	25.9	32.7	125.7
Newcastle-under-Lyme	358.2	565.9	773.5	163.1	239.4	315.6	15.0	20.8	26.6	222.6
Peterborough	263.0	335.8	408.5	74.4	129.9	185.3	8.1	11.9	15.6	123.2
Scunthorpe	129.0	263.1	397.2	63.5	107.5	151.5	7.5	11.1	14.8	101.5
Southport	91.3	160.3	229.3	48.8	92.0	135.1	4.8	8.5	12.2	81.5

Table 6 - ZINC (ug/l)

	First draw			Random daytime			Fully flushed			Intake (ug/day)
	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	mean
Ayr	120.6	138.8	157.1	87.3	108.4	129.5	19.6	25.1	30.6	99.3
Burnley	30.5	43.8	57.2	18.9	22.9	26.8	12.8	15.1	17.4	26.6
Carlisle	53.8	80.8	107.7	19.1	30.1	41.1	<6.0	<6.0	<6.0	29.3
Darlington	34.5	43.7	53.0	35.5	43.2	50.9	33.5	45.2	56.9	55.0
Dewsbury	49.9	57.9	65.9	28.7	32.2	35.8	22.2	30.8	39.3	41.5
Dunfermline	36.6	49.5	62.5	20.9	29.1	37.4	<6.0	8.8	13.9	28.6
Exeter	55.3	68.8	82.4	18.4	28.9	39.5	6.2	7.3	8.5	29.2
Falkirk	35.2	46.2	57.2	16.2	22.7	29.2	9.0	15.1	21.3	26.6
Harrogate	37.5	68.5	99.5	34.8	58.3	81.9	21.1	41.5	62.0	65.9
Merthyr Tydfil	59.6	74.2	88.8	27.9	37.8	47.7	8.6	12.6	16.6	38.3
Shrewsbury	34.9	41.7	48.6	20.5	30.8	41.2	13.2	14.3	15.3	31.7
Wigan	34.4	43.0	51.5	12.7	16.3	20.0	6.1	7.4	8.7	18.3
Bedford	45.0	63.7	82.3	22.8	30.9	39.0	<6.0	<6.0	<6.0	28.1
Gloucester	54.9	71.6	88.3	33.2	38.9	44.6	21.3	22.3	23.2	43.4
Grimsby	47.7	60.0	72.3	10.8	14.9	19.0	<6.0	<6.0	<6.0	16.6
Guildford	137.6	179.5	221.4	52.2	90.7	129.2	<6.0	12.7	19.7	83.9
Ipswich	139.5	187.0	234.5	47.5	80.3	113.0	11.7	13.6	15.4	77.5
Maidstone	91.3	173.6	255.8	37.0	62.1	87.3	<6.0	9.1	14.1	61.5
Mansfield	78.1	101.3	124.5	30.7	43.0	55.2	6.6	7.6	8.6	41.7
Newcastle-under-Lyme	39.8	55.9	72.1	17.3	25.2	33.1	<6.0	6.1	6.8	25.0
Peterborough	53.8	96.8	139.7	11.3	25.9	40.6	<6.0	<6.0	<6.0	27.5
Scunthorpe	143.7	188.7	233.7	43.1	58.2	73.2	<6.0	<6.0	6.8	58.4
Southport	30.9	54.8	78.7	19.3	27.8	36.2	<6.0	<6.0	6.2	26.5

Table 7 - IRON (ug/l)

	First draw			Random daytime			Fully flushed			Intake (ug/day)
	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	mean
Ayr	73.9	87.6	101.2	79.0	88.2	97.5	77.7	92.8	107.8	112.3
Burnley	121.3	165.2	209.1	112.2	142.4	172.6	112.6	147.1	181.6	181.9
Carlisle	49.8	141.3	232.9	83.2	114.0	144.8	77.9	105.9	133.8	140.8
Darlington	55.3	71.1	86.8	92.3	119.4	146.4	71.9	118.4	164.8	145.1
Dewsbury	94.3	135.3	176.3	86.8	130.3	173.9	87.8	123.2	158.6	160.0
Dunfermline	31.7	39.3	47.0	35.5	47.4	59.3	30.6	37.8	45.1	54.2
Exeter	46.9	72.4	98.0	50.3	68.4	86.5	46.0	74.5	103.0	88.6
Falkirk	17.1	68.6	120.2	26.6	47.2	67.8	24.8	44.3	63.8	59.3
Harrogate	165.8	196.6	227.3	182.4	212.0	241.6	183.0	210.8	238.7	263.3
Merthyr Tydfil	<4.0	209.1	430.7	76.6	94.7	112.8	78.2	96.1	114.1	127.6
Shrewsbury	12.6	17.8	23.0	18.5	24.5	30.4	15.6	23.8	32.0	29.8
Wigan	71.2	95.7	120.2	88.2	119.0	149.8	99.3	138.7	178.2	156.1
Bedford	15.6	26.5	37.3	22.2	48.2	74.2	13.1	26.4	39.7	48.5
Gloucester	9.8	16.5	23.2	16.1	22.5	28.9	10.2	21.1	32.0	27.0
Grimsby	<4.0	8.3	14.8	12.2	18.8	25.4	<4.0	4.5	7.9	16.1
Guildford	34.6	95.8	157.0	18.8	53.2	87.6	14.0	30.0	45.9	59.0
Ipswich	9.1	33.5	57.9	18.1	36.6	55.1	13.3	30.0	46.6	42.4
Maidstone	<4.0	90.9	188.8	24.6	43.6	62.7	12.9	18.2	23.5	46.3
Mansfield	16.4	47.1	77.8	37.4	50.6	63.8	36.9	49.9	62.9	62.7
Newcastle-under-Lyme	9.9	15.2	20.5	8.7	14.9	21.2	4.8	19.4	34.1	20.8
Peterborough	43.4	52.8	62.2	43.8	64.9	86.1	<4.0	88.3	201.3	91.1
Scunthorpe	16.0	25.1	34.1	14.9	27.0	39.1	13.6	33.5	53.3	36.6
Southport	9.9	24.8	39.6	26.7	34.5	42.3	19.5	25.5	31.5	38.2

Table 8 - MANGANESE (ug/l)

	First draw			Random daytime			Fully flushed			Intake (ug/day)
	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	lower 95% conf limit	mean	upper 95% conf limit	mean
Ayr	15.4	18.4	21.4	16.8	19.7	22.7	16.2	23.1	30.0	26.1
Burnley	24.2	32.7	41.2	26.4	35.1	43.8	28.0	36.6	45.1	44.4
Carlisle	11.1	22.7	34.3	14.3	18.2	22.1	13.4	16.3	19.2	22.2
Darlington	11.1	12.4	13.7	19.2	23.4	27.5	17.8	22.7	27.7	28.1
Dewsbury	28.1	36.9	45.7	27.9	39.8	51.7	29.9	42.4	54.9	50.7
Dunfermline	2.3	2.6	2.8	2.2	2.6	3.0	2.7	3.0	3.3	3.5
Exeter	3.9	4.5	5.1	5.7	6.8	7.8	4.0	8.3	12.6	9.0
Falkirk	7.8	9.3	10.7	8.7	10.1	11.4	9.8	11.1	12.4	13.0
Harrogate	57.3	64.1	70.8	59.8	66.0	72.2	59.1	65.3	71.4	82.0
Merthyr Tydfil	13.4	18.0	22.7	13.9	17.7	21.5	14.6	17.9	21.2	22.2
Shrewsbury	6.8	7.7	8.7	5.9	7.2	8.4	5.1	6.0	6.9	8.4
Wigan	6.6	7.5	8.4	6.3	10.4	14.5	5.2	10.9	16.6	13.0
Bedford	3.9	4.6	5.3	4.5	5.3	6.0	3.4	4.1	4.7	6.0
Gloucester	2.1	2.7	3.3	2.3	3.0	3.7	<2.0	2.9	4.3	3.7
Grimsby	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0	<2.0	1.3
Guildford	<2.0	2.3	3.2	<2.0	2.1	2.9	<2.0	<2.0	2.4	2.5
Ipswich	2.0	3.7	5.3	<2.0	2.9	4.2	<2.0	2.8	4.0	3.6
Maidstone	<2.0	<2.0	<2.0	<2.0	2.8	3.6	<2.0	3.0	4.2	3.4
Mansfield	<2.0	<2.0	2.3	<2.0	<2.0	2.1	<2.0	<2.0	2.3	2.2
Newcastle-under-Lyme	<2.0	2.0	2.5	<2.0	2.1	2.7	<2.0	2.0	2.5	2.6
Peterborough	<2.0	2.2	2.9	<2.0	<2.0	<2.0	7.3	9.0	10.6	5.2
Scunthorpe	4.0	5.1	6.1	<2.0	<2.0	<2.0	<2.0	<2.0	2.4	2.3
Southport	18.5	20.4	22.3	19.7	21.7	23.7	19.3	21.2	23.1	26.8

Table 9 - Service pipe and plumbing materials

	Total number of houses	Number of houses with:-			
		lead pipe	copper pipe	polythene pipe	galvanised iron pipe
Ayr	40	17	38	7	1
Burnley	40	33	37	6	0
Carlisle	38	16	37	7	0
Darlington	40	20	39	9	0
Dewsbury	39	12	31	0	0
Dunfermline	40	24	33	9	7
Exeter	39	33	36	5	0
Falkirk	39	16	38	2	0
Harrogate	40	27	38	0	1
Merthyr Tydfil	40	23	39	4	0
Shrewsbury	40	9	38	14	0
Wigan	39	23	39	11	0
Bedford	39	14	38	17	4
Gloucester	40	16	38	3	0
Grimsby	39	24	35	15	0
Guildford	23	2	20	2	4
Ipswich	40	28	37	3	2
Maidstone	37	0	25	7	30
Mansfield	38	17	37	7	0
Newcastle-u-Lyme	39	29	39	2	0
Peterborough	38	2	38	35	0
Scunthorpe	40	15	39	9	7
Southport	40	40	35	0	0

and manganese were estimated on a town-by-town basis as follows:-

$$\text{intake} = (0.06[\text{FD}] + 0.57[\text{RD}] + 0.37[\text{FF}]) \times 1.25\ \mu\text{g day}^{-1}$$

where [FD], [RD] and [FF] are the town mean concentrations in μgl^{-1} of the first draw, random daytime and fully-flushed samples and 1.25 is the estimate of the mean total daily volume of water consumed by typical middle-aged men in Britain. These exposure estimates are shown for the 5 metals in the final column of Tables 4 - 8. In the case of lead (Table 4) these estimates are based only on houses where lead pipe is present somewhere in the service connections or plumbing.

4.4. CHANGES IN TRACE ELEMENT AND MAJOR ION LEVELS AFTER PROLONGED STAGNATION

Table 10 gives the results of a paired comparison of trace element and major ion levels between first drawn and fully-flushed samples for the group of 12 soft water towns and the group of 11 hard water towns. The results are expressed as percentage mean differences between the first-draw and fully-flushed concentrations; a negative value indicating a decrease in concentration. The number of observations used to form the means is also shown. The statistical significance of the concentration changes was tested using the non-parametric Wilcoxon matched-pairs signed-ranks test(6) instead of the "t"-test. This test was preferred to avoid making the assumptions of normality required for the "t"-test and, as a ranking method, it was less sensitive to the effect of outlying observations. The Wilcoxon test rejected values that were the same in both the first-draw and fully-flushed samples and in the majority of cases this occurred where both samples in the pair were below the criterion of detection. Data rejected in the significance test were also rejected when forming the percentage mean differences shown in Table 10.

4.5. MUTLIPLE REGRESSION ANALYSIS

A multiple regression analysis revealed which of the major water quality parameters and trace metals were most important in controlling the levels of plumbing related metals in tap-water. The analysis was confined to the three most important plumbing related materials: copper, lead and zinc. First-draw concentration values were used in the analysis as these correspond most closely to the maximum equilibrium metal concentration obtainable after prolonged contact of water with the service pipe and household plumbing.

As some major water quality parameters are related to source of supply

Table 10 - Comparison of first draw and fully flushed trace element and major ion concentrations

	SOFT WATER TOWNS		HARD WATER TOWNS	
	% mean difference in concentration	number of observations~	% mean difference in concentration	number of observations~
As	0.2	98	-4.9	90
Al	6.2	452	-5.2	196
B	19.2***	397	2.8**	379
Ba	3.7***	411	4.3***	384
Be	-19.0*	72	14.0	35
Bi	-45.0***	90	-45.2*	96
Ca	1.7***	464	1.4***	423
Cd	40.4**	22	17.1	13
Co	-10.2	45	-2.8	28
Cr	5.5	201	37.1	110
Cu	502.1***	447	2051.8***	422
Fe	7.5***	458	26.7	376
K	5.1***	462	1.7***	422
Li	5.5*	245	-5.9	341
Mg	1.9***	463	2.1***	423
Mn	-10.4***	408	-9.9	231
Mo	1.6*	201	-22.0***	246
Na	2.6***	464	1.8	423
Ni	16.4	301	27.3	193
Pb	313.4***	350	391.5***	254
Si	2.4***	463	1.6***	422
Sr	2.9***	370	2.3***	408
Ti	1.6	284	6.7	388
V	13.7***	283	-3.4	363
Zn	232.9***	462	1230.9***	423
Zr	11.1*	124	-4.8	183

~ The number of observations refers to those included in the test of significance. All values that were the same in both samples were rejected

* Difference significant at the 5% level
** Difference significant at the 1% level
*** Difference significant at the 0.1% level

rather than town, the means of the house data corresponding to individual water sources within the towns were used in preference to the town means or the individual house data. A total of 64 sources were identified supplying the 23 survey towns. Missing data (mainly pH data) reduced the numbers to 49 sources in 20 towns and the mean values of the various water quality parameters of interest in these 49 sources formed the basic data base for the multiple regression analysis.

A multiplicative model of the general form:-

$$\log_e[\text{metal}] = \alpha + \beta\text{pH} + \sum_i \gamma_i \log_e x_i + \varepsilon$$

was selected for all the multiple regression analyses as it was found that this type of model explained more of the total variance about the means of y and gave much better residual error structures than any of the other types of model tried.

4.5.1. Copper

Initially, four independent variables were included in the model for copper, these were: pH, total hardness, alkalinity and temperature of flushed tap-water. The temperature of tap-water was found not to have a significant effect at the 5% level and total hardness and alkalinity were so highly correlated ($r = 0.94$) that their effects could not be separated in a single model. The final models for copper were, therefore:-

i) $\log_e[\text{Cu}] = 13.3 - 1.3\text{ pH} + 0.27 \log_e [\text{total hardness}]$

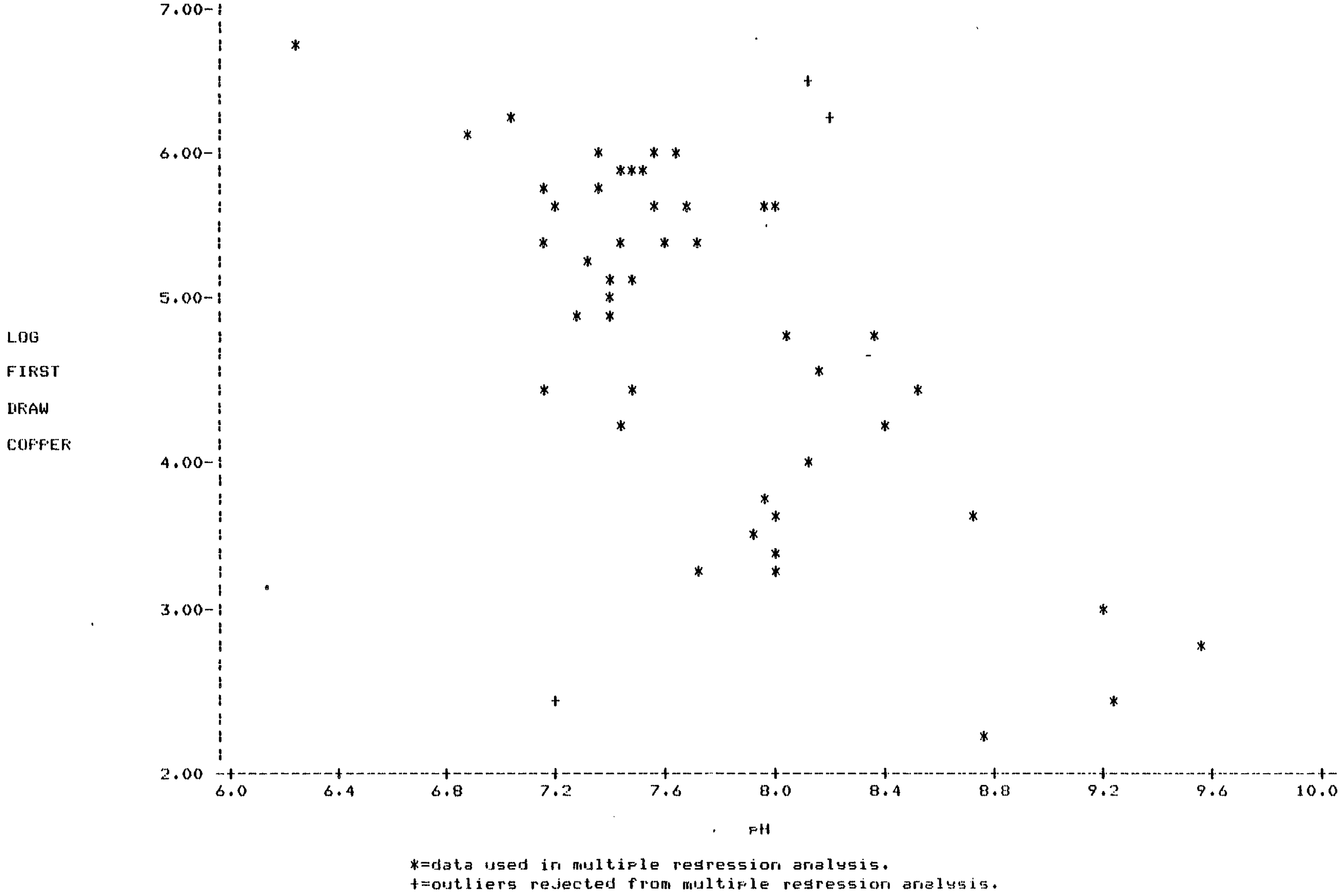
ii) $\log_e[\text{Cu}] = 13.6 - 1.2\text{ pH} + 0.22 \log_e [\text{alkalinity}]$

with pH as the major explanatory variable in each case. Both models explained 63% of the total variation about the mean copper concentration. The residuals histogram revealed 3 main outliers which were excluded from the final analysis. It was not possible to find specific reasons for these outliers but the on-site measurement of pH can present instrumentation problems so that the outliers were tentatively attributed to faulty pH readings. Another possibility is wrong allocation of houses to sources. A scatter plot of $\log_e[\text{Cu}]$ against pH is shown in Figure 2 with the three outlying observations identified.

4.5.2. Lead

Initially, seven independent variables were included in the model for lead,

Figure 2 - TO SHOW RELATIONSHIP BETWEEN FIRST DRAW COPPER AND pH.



these were: pH, total hardness, alkalinity, tap-water temperature, chloride, silicon and copper. Temperature, chloride, silicon and copper did not exert significant effects at the 5% level and the intercorrelation between total hardness and alkalinity meant that their effects could not be separated in the model; as was the case for copper (Section 4.5.1). Alkalinity was selected for inclusion in the final model in preference to total hardness because it was considered to be more relevant to the chemistry of lead corrosion. Inclusion of the data for all 49 sources in the initial model produced a very poor residual error structure so the soft and hard water sources were separated and all houses known not to have lead service pipes or plumbing were excluded. After exclusion of missing data, there remained mean values for 20 soft water sources and 21 hard water sources.

The final model for lead in soft water towns was:-

$$\log_e[\mathrm{Pb}] = 9.2 - 0.57\ \mathrm{pH} - 0.32 \log_e [\mathrm{alk}]$$

with pH as the major explanatory variable (alkalinity was significant only at the 10% level). The model explained 31% of the total variation about the mean lead concentration. A scatter plot of $\log_e[\mathrm{Pb}]$ against pH is shown in Figure 3. Examination of the residuals revealed no serious outliers.

No model could be fitted for hard water towns as none of the independent variables significantly reduced the residual variance.

4.5.3. Zinc

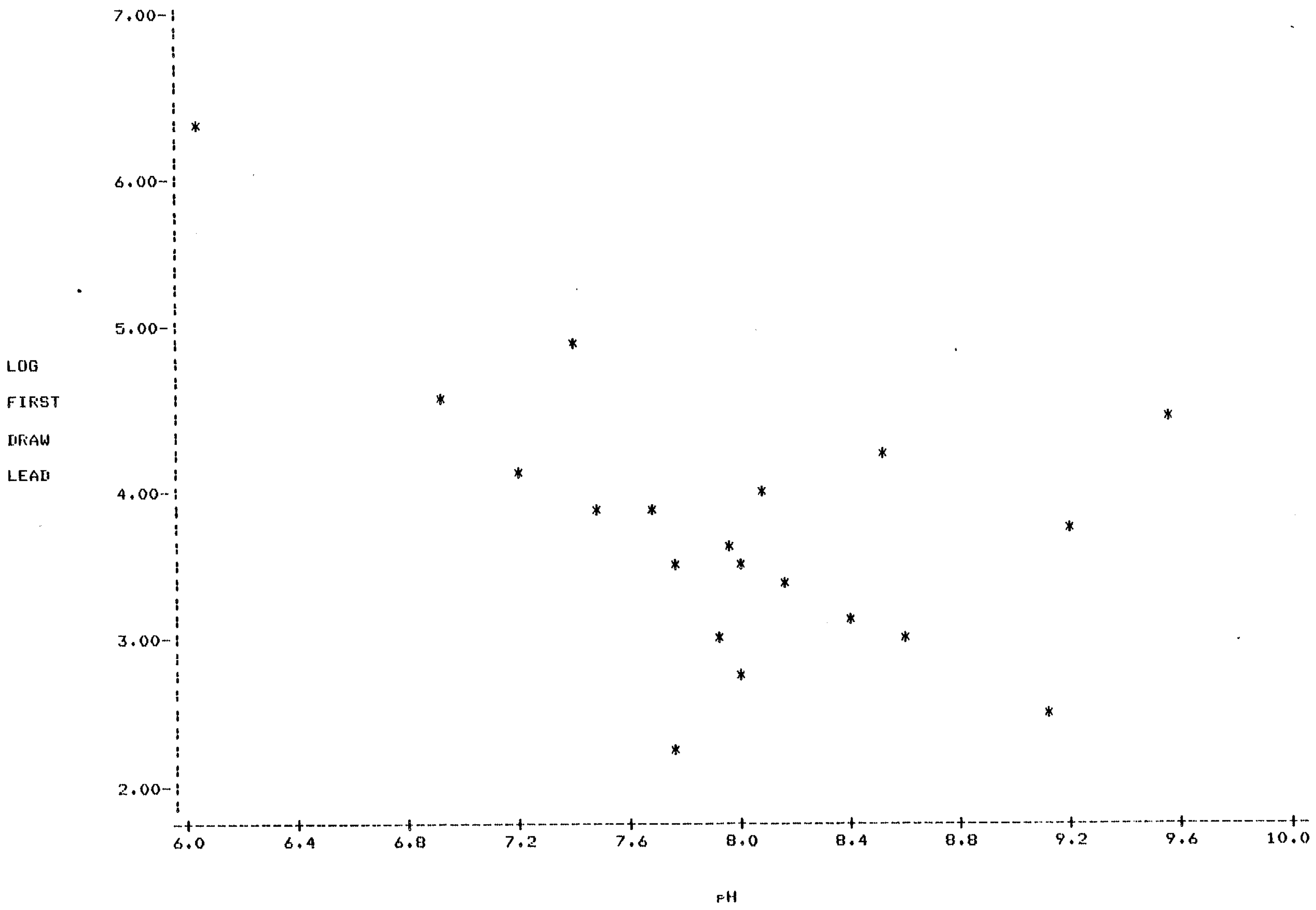
Seven independent variables were included in the initial model for zinc. These were the same as those included in the inital model for lead (Section 4.5.2) plus the chloride/alkalinity ratio (related to dezincification propensity). Only pH and first-draw copper were found to exert significant effects at the 5% level but since these two variables were highly intercorrelated ($r = -0.63$) (Section 4.5.1) their separate effects could not be resolved adequately in a single model. The final models for zinc were, therefore:-

i) $\log_e[\mathrm{Zn}] = 7.8 - 0.44\ \mathrm{pH}$

ii) $\log_e[\mathrm{Zn}] = 3.1 + 0.27 \log_e [\mathrm{Cu}]$

Model i) explained 23% and model ii) explained 29% of the total variation

Figure 3 - TO SHOW RELATIONSHIP BETWEEN FIRST DRAW LEAD AND pH.



about the mean zinc concentration respectively. Examination of the residuals revealed 3 main outliers which were excluded from the final analysis. One of these outliers was the same observation identified as an outlier in the copper model (section 4.5.1). Again no specific reasons for these outliers could be found but the general reasons given for the copper outliers probably also apply to the zinc data. Scatter plots of $\log_e[\mathrm{Zn}]$ against pH and $\log_e[\mathrm{Zn}]$ against $\log_e[\mathrm{Cu}]$ are shown in Figures 4 and 5 respectively with the outlying observations identified.

5. DISCUSSION

5.1. DRINKING WATER CONSUMPTION HABITS

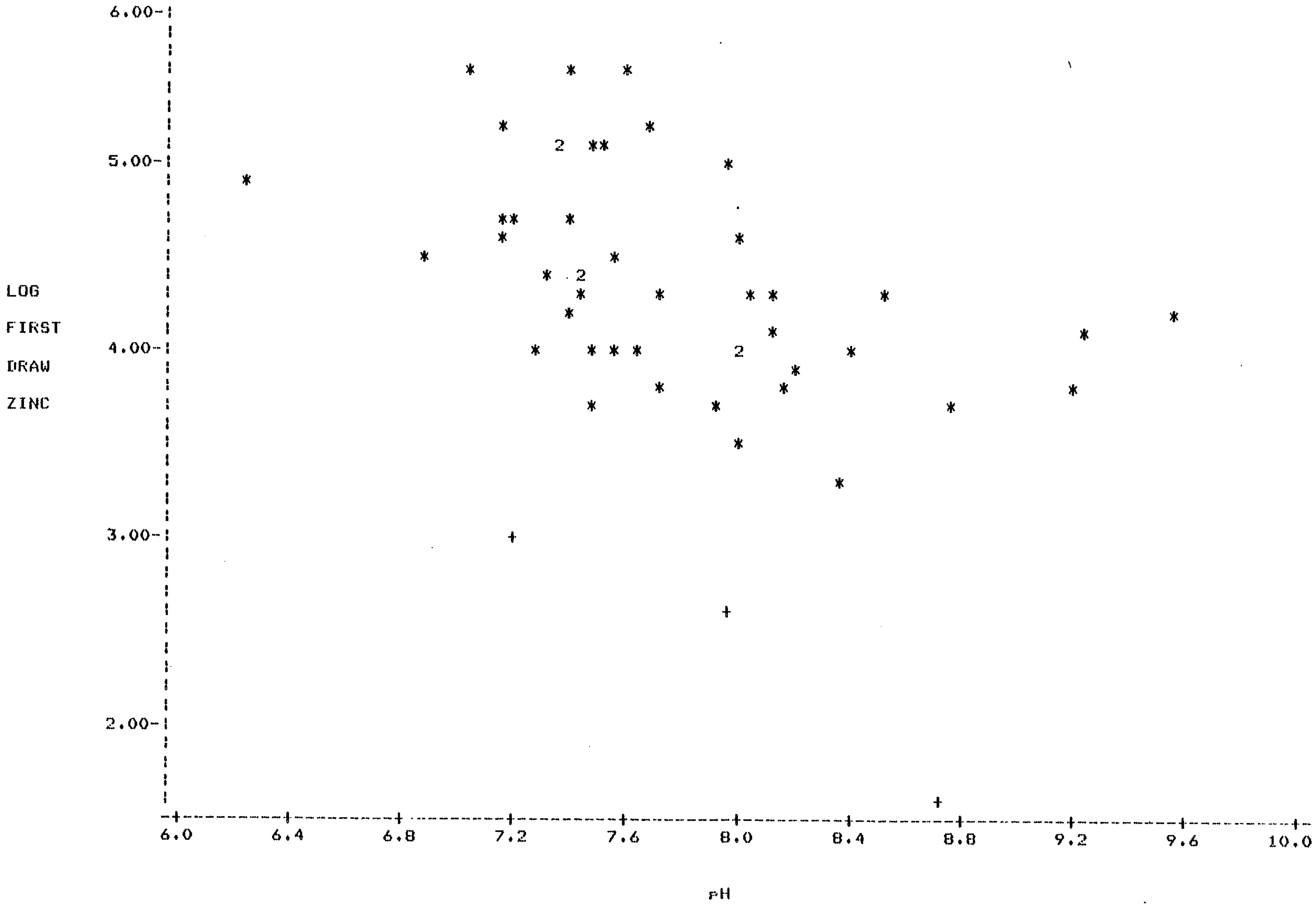
From the water consumption questionnaire, based on the results for all 1099 subjects in the 25 towns, the mean tap-water intake was estimated to be 1.25 litres per day. Tea accounts for 68% of this, and coffee 20%. Taps in the home accounted for 76% of the tap-water intake, 95% of this being from the kitchen cold tap and 5% from the hot tap. Of the home consumption, 57% was estimated to be in the 'random daytime' category, i.e. taken as it comes. The 'flushed' category accounted for 37% and 6% was 'first-draw' water.

For comparison, information from the separate WRC survey(7) was analysed for men of the same age group (40 - 59), 420 in all. Results for this were in close agreement, the total consumption being 1.20 litres per day, 70% being tea and 19% coffee. Home taps provided 72% of the water with 92% from the kitchen tap and 7% from the hot tap. The main discrepancy was in the allocation of volumes of water consumed between 'random daytime' and 'flushed': 83% and 13% respectively in the separate survey. The first draw estimate, at 4% was in close agreement. If the above allocations are used to calculate the mean daily intakes of trace metals shown in Tables 4 - 8 (Section 4.2.3), the estiamtes are increased approximately between 6 and 25%,

5.2. TRACE METAL EXPOSURE ESTIMATES

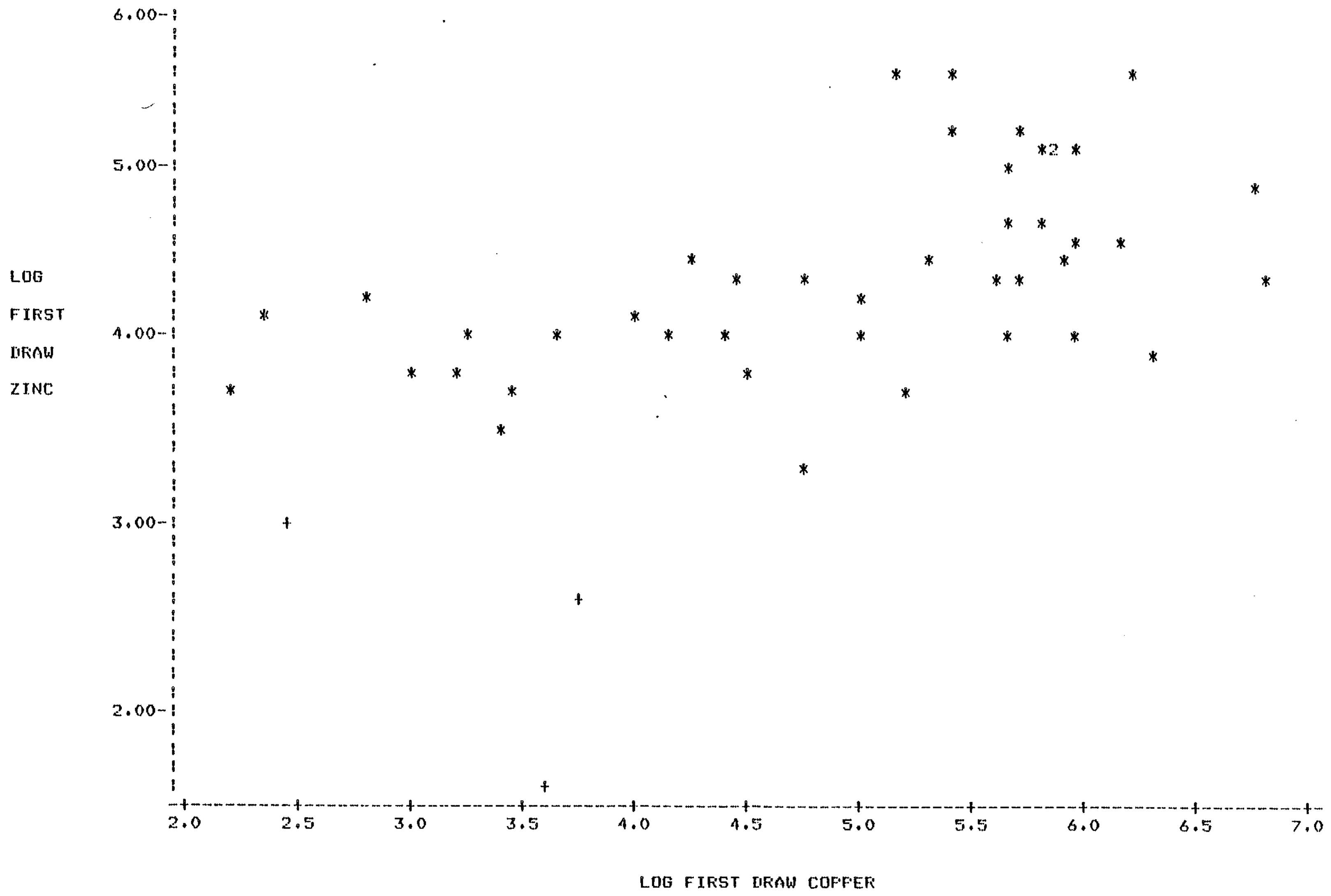
Estimates of population exposure in the form of predicted mean daily intakes were calculated on a town-by-town basis for the five most important plumbing and distribution system related trace metals, i.e. lead, copper, zinc, iron and manganese. The method used to predict these intakes is described fully in Section 4.3. It should be noted that the mean daily volume of water consumed was estimated over all 25 towns and not on a town-by-town basis. This approach has the advantage of smoothing out seasonal fluctuations in

Fisure 4 - TO SHOW RELATIONSHIP BETWEEN FIRST DRAW ZINC AND pH.



*=data used in multiple regression analysis.
(where numbers appear=more than one point)
+=outliers rejected from multiple regression analysis.

Figure 5 - TO SHOW RELATIONSHIP BETWEEN FIRST DRAW ZINC AND FIRST DRAW COPPER.



*=data used in multiple regression analysis.
(where numbers appear= more than one point)
+=outliers rejected from multiple regression analysis.

drinking water consumption habits as the towns were sampled at different times of the year (Table 2).

Tables 4 to 8 show the predicted mean intakes for the five metals together with estimated town mean concentrations for the 3 sample categories. In each table the 12 soft water towns (Ayr - Wigan) appear first, followed by the 11 hard water towns (Bedford - Southport). Some of the main points emerging from these tables are as follows:-

i) Mean lead and copper exposure levels for Ayr greatly exceed those for other towns. The very soft, acidic (no pH correction is applied) supply to Ayr is highly corrosive to lead and copper plumbing. In addition, there is a high proportion of lead service connections in Ayr.

ii) With the exception of Ayr, copper exposure levels are generally higher in the hard water towns than in the soft water towns.

iii) Iron and manganese exposure levels are generally higher in soft water towns than in hard water towns.

Lack of space precludes giving town-by-town statistical summaries for the remaining 21 trace elements and major ions determined in the survey. Table 3 gives a comparison between hard and soft water towns for these parameters.

5.3. CHANGES IN TRACE ELEMENT AND MAJOR ION LEVELS AFTER PROLONGED STAGNATION

Any elements or ions present in water that are unaffected by contact with service pipes or household plumbing will have similar levels in first-draw and fully-flushed samples. However, elements or ions which either leach out or deposit on contact with pipes will have different levels in the two samples. The magnitude and direction of the statistically significant concentration differences between the two sample categories are discussed below for various groups of elements and ions.

i) Lead, copper and zinc

As expected, these 3 metals show very large increases in first-draw samples compared with flushed samples for both hard and soft water towns (Table 10). For lead the magnitude of the increase was similar in both hard and soft water towns whilst for copper and zinc the increase in hard water towns was much greater. In the case of zinc, this could reflect the greater use of galvanised iron services in hard water towns. For lead the results apply to only those houses with lead pipes present in the service connections or plumbing so that the statistical populations being compared between hard and soft water towns comprise houses with lead pipes.

ii) Boron, barium, calcium, potassium, lithium, magnesium, sodium silicon and strontium.

These 9 elements were not expected to change after prolonged contact with the service pipes and plumbing. However most showed slight but statistically significant increases in the first draw samples (the exception was boron in the soft water towns with an increase of 19%).

In all cases the slight increases observed (around 1 to 5%) were attributed to a sampling artefact caused by the addition of a smaller volume of preserving acid to the first-draw samples compared with the random daytime or flushed samples. The resulting extra dilution of the random daytime and flushed samples was expected to be around 2%.

iii) Beryllium, bismuth, manganese and molybdenum

These 4 elements show significant decreases in first-draw samples suggesting that they are deposited or adsorbed on prolonged contact with the pipes. Beryllium and manganese only show significant decreases in soft water towns and molybdenum only in hard water towns.

It has long been recognised that manganese levels tend to decrease in distribution systems due to adsorption onto precipitated mains deposits(8,9,10). Presumably similar mechanisms apply in house service pipes. Manganese levels are considerably greater in soft water towns (Table 3) and in both soft and hard water towns, the mean levels are well above the analytical limit of detection. In the cases of beryllium, bismuth and molybdenum the group means lie below the limit of detection.

iv) Cadmium, iron, vanadium and zirconium

These 4 metals show statistically significant increases in the first-draw samples in soft water towns. It can be seen from the number of observations (Table 10) that the 40% increase in cadmium concentration was observed in only 22 samples in which cadmium was detected. However, this does show that cadmium is leached out on prolonged contact of water with pipes in the relatively small number of cases where it is present in detectable amounts.

The remaining elements: silver, aluminium, cobalt, chromium, nickel and titanium do not show statistically significant changes in concentration after prolonged contact with pipes.

5.4. MULTIPLE REGRESSION ANALYSIS

The aim of the multiple regression analysis was to reveal which water quality parameters and trace elements have a major effect on the concentration of copper, lead and zinc in drinking water. This is all the information that can be brought out from this kind of survey data.

The main points to emerge from the analysis was that pH was the most important factor influencing equilibrium levels of lead and copper in tap-water and that pH and copper concentration were almost equally important in influencing equilibrium zinc levels. Alkalinity or total hardness (these variables were so highly intercorrelated that their effects were indistinguishable) also had a significant effect on lead and copper levels, although for lead the level of significance was only 10%.

5.4.1. Copper

The results for copper support previous studies(11) which have shown that copper solvency is highly pH dependent in both soft and hard waters. The correlation between pH and total hardness/alkalinity is reasonably low ($r = 0.27$) showing that there is only a slight tendency for hard waters to be associated with higher pH values.

5.4.2. Lead

The results for lead show pH dependency only in soft (low alkalinity) waters. This conclusion supports the recent work at WRC on predicting drinking water lead levels using theoretical solubility calculations(12). These calculations showed that lead is expected to be highly pH dependent in low alkalinity waters (100 mg $CaCO_3$ l^{-1}) whilst alkalinity itself is predicted to have relatively little direct effect on lead solubility. In high alkalinity waters (100 mg $CaCO_3$ l^{-1}) pH dependence is not theoretically predicted.

5.4.3. Zinc

As zinc is a highly electronegative metal its corrosion is expected to be enhanced by the deposition of more electropositive metals like copper.

Although the multiple regression analysis shows a strong positive association between zinc and copper levels, this could merely reflect the strong negative association between copper and pH (Section 5.4.1). The multiple regression analysis reveals the well documented(11) negative association of zinc with pH. There are two distinct sources of zinc in tap-water: corrosion of the zinc coatings of galvanised service pipes and dezincification of brass plumbing fittings. It is outside the scope of the present analysis to attempt to assess the relative importance of the two sources.

6. CONCLUSIONS

i) Water consumption habits for the middle-aged men involved in the Regional Heart Study were very similar to those recorded for men of the same age group in the recent WRC survey of drinking water consumption in Great Britain. Results from both studies show that the average daily consumption of tap-water for this age group (40 - 59) is approximately 1.2 litres per head per day.

ii) Estimates of population exposure levels of middle-aged men to trace elements in drinking water were derived by calculating mean daily intakes from information on tap-water trace element concentrations combined with information on daily water consumption.

iii) Copper and zinc levels increase on prolonged contact with service pipes to a greater extent in hard water areas than in soft water areas. Where lead pipes are present, lead levels increase in similar proportion in hard and soft water areas after prolonged contact with service pipes. Although cadmium is infrequently detected in tap-water and its levels are very low, concentrations in soft water areas are significantly increased on prolonged contact with service pipes.

iv) Manganese levels decrease slightly in soft water areas after prolonged contact with service pipes.

v) With the exception of iron, vanadium and zirconium, which show slight increases in soft water areas, no other trace elements appeared to show important changes in concentration on prolonged contact with service pipes.

vi) The most important of the measured water quality factors controlling tap-water copper levels is pH. Alkalinity (or total hardness) has a slight positive association with tap-water copper levels.

vii) The most important factor controlling tap-water lead levels in soft waters is pH. Alkalinity has a slight negative association with tap-water lead levels. No pH dependence was observed for lead in hard waters.

viii) pH and copper concentration appear to be equally important in controlling tap-water zinc levels. There is a strong positive association between zinc and copper levels in tap-water but this could be reflecting the strong intercorrelation between tap-water pH and copper levels.

REFERENCES

1. SHAPER, A.G., POCOCK, S.J., WALKER, MARY, COHEN, N.M., WALE, C.J., and THOMPSON, A.G. British Regional Heart Study: Cardiovascular risk factors in middle-aged men in 24 towns. Brit. Med. J., July 1981, 283, 79-186.

2. NORTON, R.L., and ORPWOOD, J.R. Multi-element analysis using an inductively-coupled plasma spectrometer. Part 1: Commissioning and preliminary evaluation of performance. WRC Technical Report TR 141, 1980.

3. BRITTON, A., and RICHARDS, W.N. A pot-pourri on plumbosolvency. Paper presented at the IWES Symposium on lead in drinking water, Birmingham, 1980.

4. OLIPHANT, R.J. Contamination of potable water by materials used in plumbing fittings. WRC Technical Report. (In preparation).

5. LYON, T.D.B., and LENIHAN, J.M.A. Corrosion in solder jointed copper tubes resulting in lead contamination of drinking water. Br. Corrosion J., 1977, 12, 41-45.

6. SIEGEL, S. NON-PARAMETRIC STATISTICS FOR THE BEHAVIOURAL SCIENCES. Ch. 5, McGraw-Hill, 1956.

7. HOPKIN, S.M., and ELLIS, J.C. Drinking water consumption in Great Britain. WRC Technical Report TR137, 1980.

8. ZOETEMAN, B.C.J., and HARING, B.J.A. The nature and impact of deterioration of the quality of drinking water after treatment and prior to consumption. Netherlands National Institute for Drinking Water Supply Contract Report No. 76-14, 1975.

9. DANGEL, R.A. Study of corrosion products in the Seattle Water Department's Tolt distribution system, USA Environmental Protection Agency Report EPA-670/2-75-036, 1975.

10. BAILEY, R.J. and RUSSELL, P.F. Trace Elements in Drinking Water. WRC Conference on Water Distribution Systems held at Keble College, Oxford, September 1978. Papers and proceedings published November 1978.

11. CAMP, T.R. WATER AND ITS IMPURITIES, Ch. 8, Reinhold Publishing Co. (1963).

12. JACKSON, P.J. and SHEIHAM, I. Calculation of lead solubility in water. WRC Technical Report TR152 (1980).

APPENDIX 2A

FACTORS AFFECTING LEAD LEVELS IN DRINKING WATER

CONTENTS

1. INTRODUCTION

Because background lead levels in the mains water supply are usually negligible$^{(1)}$, domestic lead service pipes and plumbing are the major sources of lead in drinking water. It has long been recognised that variations in contact time of water with lead service pipes and plumbing, resulting from normal daily household water use patterns, give rise to a high degree of within-house variation in drinking water lead levels$^{(2-6)}$. Although this variation is mainly due to the increase in lead concentration with increasing contact time in the pipes, other factors such as the flow-rate at which water is drawn from the tap, the rate of opening the tap and the volume of water drawn in relation to the length of pipe also contribute to the variation in lead levels.

The overall aim of the work described in this Appendix is to assist the development of efficient and economical sampling methods for the estimation of lead levels in drinking water, in particular for estimating population exposure to lead.

Experiments were designed to answer the following specific questions:-

(i) How does flow-rate affect the concentration of lead in tap-water when a lead service pipe is continuously flushed to reach steady state conditions?

(ii) How does the stagnation time of water in the service pipe affect the concentration of lead in tap-water? Can the relationship between stagnation time and lead concentration be described by a simple mathematical model?

(iii) Does the flow-rate at which stagnation samples are taken have any effect on the lead concentration?

(iv) Does the proportion of insoluble lead material in tap-water vary with flow-rate?

2. EXPERIMENTAL METHODS

Three *in situ* lead pipes were selected for the study: one in South-East England (pipe A) and two in Scotland (pipes B and C). The main criterion in the selection of the pipes was that they were supplied with two completely different types of water, both known to produce high lead levels in drinking water. Pipe A was fed from a hard borehole source (typical total hardness = 300 mg $CaCO_3\ l^{-1}$) with high carbonate hardness and fairly low pH (pH ≃ 7.2). Pipes B and C were supplied from *the same* very soft, acidic upland catchment source (typical total hardness = 12 mg $CaCO_3\ l^{-1}$, pH ≃ 6.3). Lime treatment was applied at source to counteract the highly plumbosolvent nature of the raw water and this resulted in typical drinking water pH values of around 7.8(7). All three pipes were located at little used Water Authority pumping stations and had been in service for many years. Details of the pipes and typical water quality analyses are given in Table 1.

Table 1. Details of *in situ* lead pipes and typical water analyses

	Pipe A	Pipe B	Pipe C
estimated length	24 metres	47 metres	16 metres
estimated bore	9.5 mm (3/8 in.nom)	13 mm (½ in.nom)	19 mm (¾ in.nom)
calculated volume	1.7 litres	6.2 litres	4.5 litres
approximate age	42 years	60 years	53 years
total hardness	311 mg $CaCO_3\ l^{-1}$	14 mg $CaCO_3\ l^{-1}$	
alkalinity	213 mg $CaCO_3\ l^{-1}$	< 5 mg $CaCO_3\ l^{-1}$	
calcium	112 mg Ca l^{-1}	4.2 mg Ca l^{-1}	
magnesium	8 mg Mg l^{-1}	0.8 mg Mg l^{-1}	
pH	7.2 units	7.8 units	
free carbon dioxide	40 mg l^{-1}	< 2 mg l^{-1}	
conductivity	603 μ mhos cm^{-1}	45 μ mhos cm^{-1}	
total organic carbon	1.9 mg C l^{-1}	1.4 mg C l^{-1}	
nitrate	5.5 mg $NO_3\ l^{-1}$	0.6 mg $NO_3\ l^{-1}$	
chloride	33 mg Cl l^{-1}	6.9 mg Cl l^{-1}	
potassium	19.5 mg K l^{-1}	0.3 mg K l^{-1}	
sodium	19.5 mg Na l^{-1}	2.8 mg Na l^{-1}	
sulphate	71 mg $SO_4\ l^{-1}$	3.9 mg $SO_4\ l^{-1}$	
silica	32 mg $SiO_2\ l^{-1}$	1.3 mg $SiO_2\ l^{-1}$	

2.1. EXPERIMENTS UNDER FLUSHING CONDITIONS

These experiments were designed to discover the relationship between lead concentration and flow-rate when the tap was continuously flushed until steady state conditions were assumed to have been reached. A 360° protractor and pointer assembly, attached to the tap, was used to indicate the extent of the tap opening. The tap angle was adjusted to obtain the required flow-rate and a period of time equivalent to flushing five pipe volumes was allowed in order to ensure complete replacement of water before the sample was taken. The tap flow-rate was estimated by measuring the volume of water collected over a 6 or 12 second period, both immediately before the sample was taken and immediately afterwards. The mean of the two measurements was taken as the typical flow-rate value during sampling. Flow-rates were varied systematically in cycles of opening and closing the tap.

2.2. EXPERIMENTS UNDER STAGNATION CONDITIONS

These experiments were designed to discover the relationship between lead concentration and stagnation time of water in the pipe and also to find out whether the lead concentrations in stagnation samples were influenced by sampling flow-rate. In the first set of experiments, samples were taken from the tap at a constant flow-rate after various stagnation times. No water was flushed before collecting the sample. The volume of sample collected was calculated beforehand to ensure that it did not exceed half the estimated volume of the pipe. Before each stagnation period at least five pipe volumes of water were flushed through the pipe to return to the steady state 'flushing base level' concentration. Samples were taken at the end of these flushing periods, just before turning off the tap, to determine the 'base level' lead concentrations at intervals during the experiment.

To determine the effect of flow-rate on the lead concentration in stagnation samples a separate experiment was devised in which samples were taken at various flow-rates after a constant stagnation period.

The effect of the rate of opening the tap on lead levels was not studied because of the problems in measuring this factor. However, efforts were made to keep the rate of the tap opening constant during the experiments in order to minimise possible effects from this source.

2.3. DETERMINATION OF TOTAL LEAD CONCENTRATION

The samples, collected in acid washed poly(ethylene) bottles, were acidified to pH 1 with 'Aristar' nitric acid in order to convert all the lead to soluble form and to prevent adsorption onto the internal surface of the bottles.

Hard water samples with concentrations less than 200 $\mu g\ l^{-1}$ were analysed by flameless atomic absorption spectrometry using a Perkin-Elmer 460 instrument equipped with HGA 76 electrothermal atomiser. Using a standard additions technique of calibration, the detection limit* was 2 $\mu g\ l^{-1}$ and the precision† at 50 $\mu g\ l^{-1}$ was estimated to be ± 5 $\mu g\ l^{-1}$ (95% confidence level).

Hard water samples with lead concentrations greater than 200 $\mu g\ l^{-1}$ were analysed by flame atomic absorption spectrometry using a Perkin-Elmer instrument.

Soft water samples were analysed by flame atomic absorption spectrometry using a Varian 175 instrument. The detection limit was 50 $\mu g\ l^{-1}$ and the precision‡ at 100 $\mu g\ l^{-1}$ was estimated to be ± 9 $\mu g\ l^{-1}$ (95% confidence level).

2.4. DETERMINATION OF NON-FILTRABLE LEAD CONCENTRATION

Samples were collected in 1 litre poly(tetrafluoroethylene) (PTFE) beakers and filtered under vacuum through 'Nuclepore' polycarbonate membrane filters of 0.4 micron pore size. The membrane filters were mounted in a holder constructed entirely of PTFE to minimise adsorption losses during filtration(8). The filtrate was collected directly into sufficient 'Aristar' nitric acid to bring the final pH of the filtrate to 1. The residue was stripped from the membrane filters by soaking them in a known volume of 0.1N 'Aristar' nitric

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* Detection limit is defined as $2\sqrt{2}$ ts, where s is an estimate of the within-batch standard deviation of the blanks with n degrees of freedom, and t is the tabulated value of Students 't' statistic (single sided) at the 0.05 probability level for n degrees of freedom.

† Precision was derived from 20 replicate determinations of a 50 $\mu g\ l^{-1}$ quality control standard.

‡ Precision was derived from 57 replicate determinations of a 100 $\mu g\ l^{-1}$ quality control standard.

acid at room temperature for at least one month. The filtrate and residue solutions were analysed by atomic absorption spectrometry as described in Section 2.3.

Adsorption losses in the PTFE filtration system were assessed using the technique of double filtration in which the *unacidified* filtrate was re-filtered, under identical conditions, through a second 0.4 micron 'Nuclepore' membrane. Since all non-filtrable material was removed in the first filtration, the difference in concentration between the first filtrate and the second filtrate gave an indication of the loss of filtrable lead in the system. In all cases, for both the hard and soft waters, this loss did not exceed 10%. However, when recoveries were assessed by adding the filtrable and non-filtrable fractions and comparing this with the total lead concentration, it was found that system losses were often considerably greater than 10%. A possible explanation for these losses is incomplete stripping of the non-filtrable residue from the membrane filters.

3. RESULTS AND DISCUSSION

3.1. EXPERIMENTS UNDER FLUSHING CONDITIONS

Pipe A (hard water)

The results of two separate experiments conducted on this pipe are shown in Fig. 1. No water had been flushed through the pipe for a period of about three months before the first experiment. Before the second experiment, water had been allowed to flush continuously through the pipe (at 4 $l\ min^{-1}$) for a period of about three weeks. In both cases there was a clear decrease in lead concentration with increasing flow-rate in the low moderate flow-rate region (0.5-3 $l\ min^{-1}$), due to decreasing contact time of the water with the internal surface of the pipe. At flow-rates above about 7 $l\ min^{-1}$ there was an upturn in lead concentration. This was much more pronounced in the first experiment, but in both cases it was significant at the 0.1% level. There was no significant difference between any of the repeat cycles in either experiment.

Pipes B and C (soft water)

The results are shown in Fig. 2. Only a small amount of water had been flushed at a moderate flow-rate through each pipe before the start of

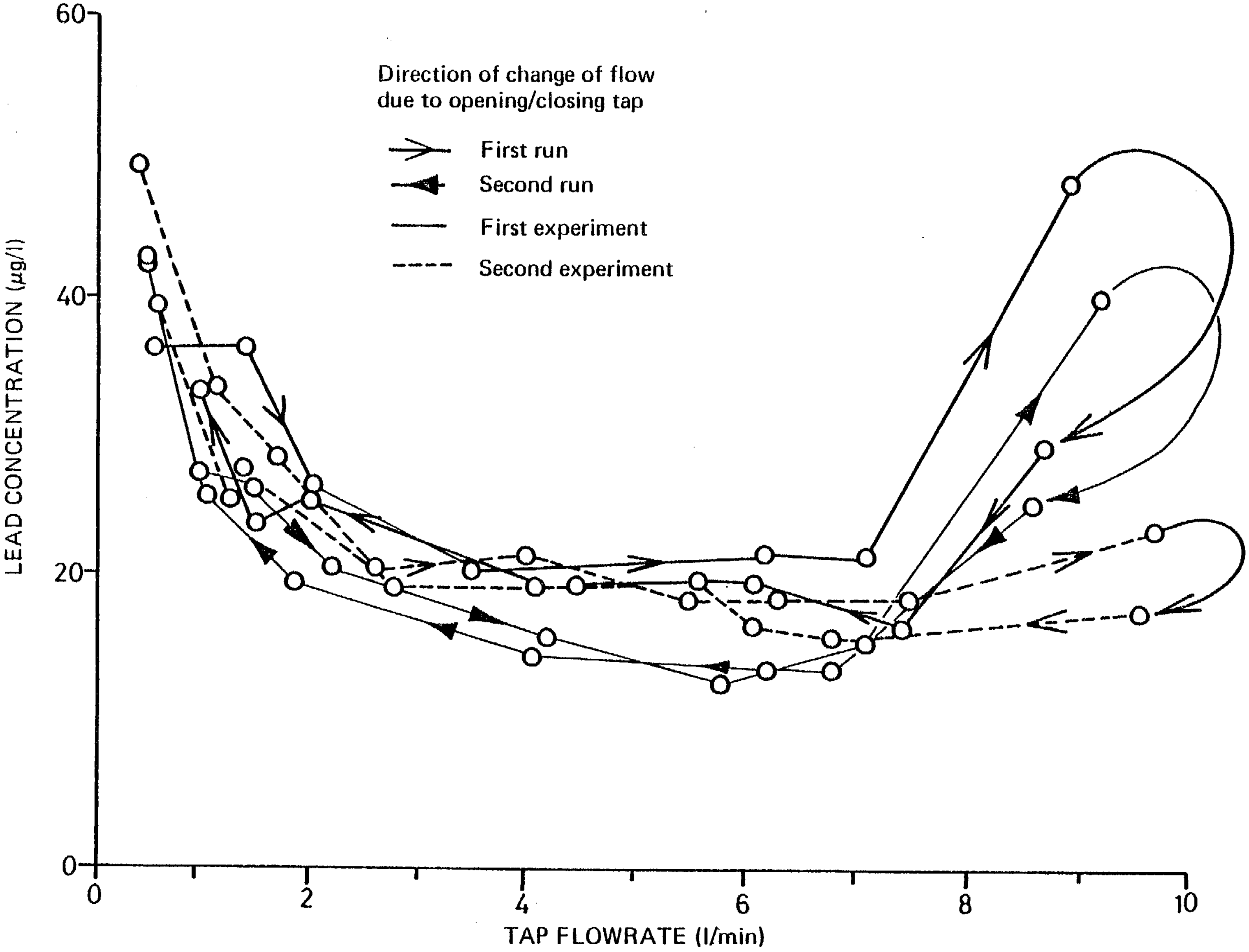

Fig. 1. To show the relationship between steady state lead concentration and flow-rate under flushing conditions – Pipe A (hard water)

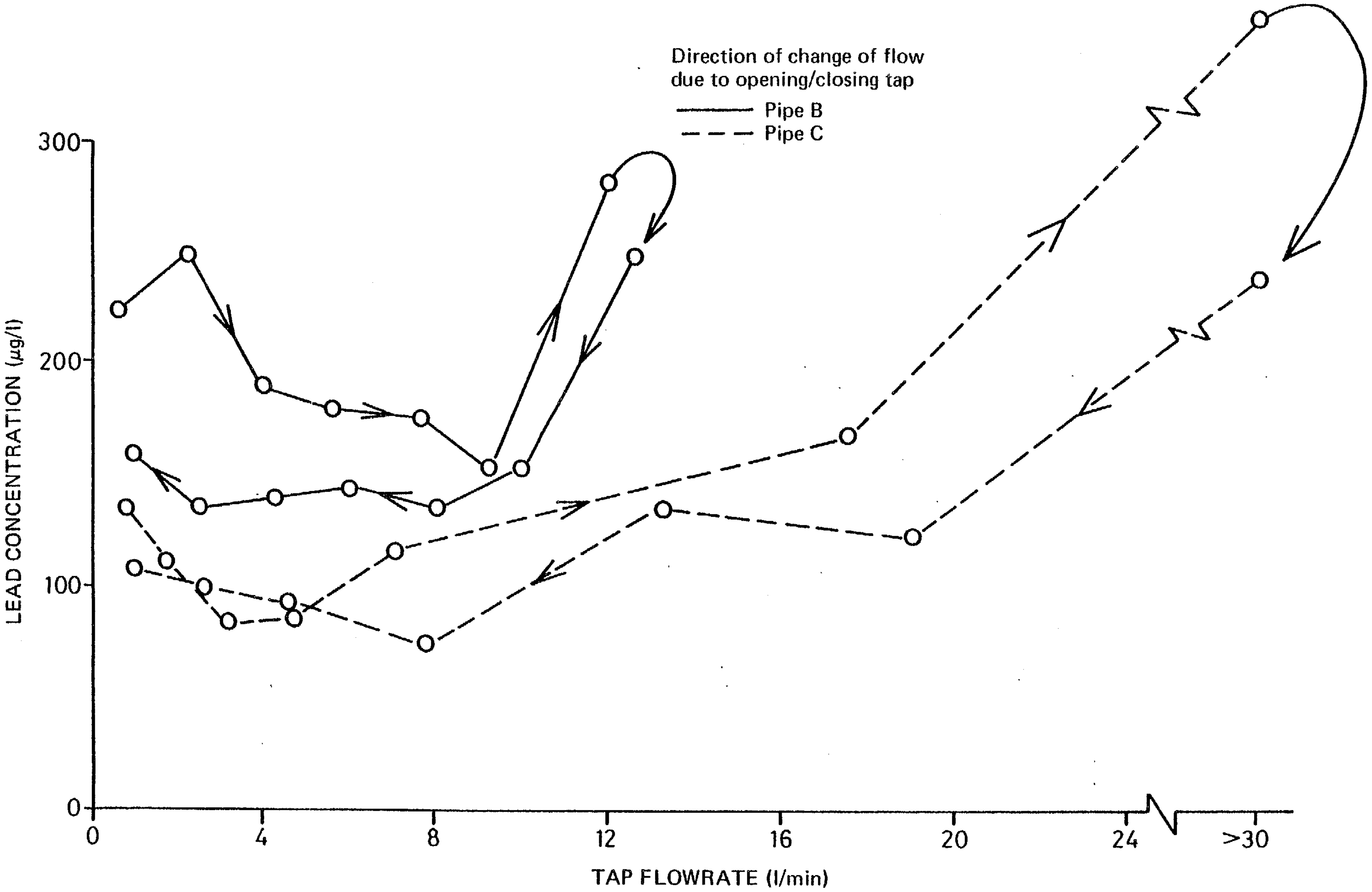

Fig. 2. To show the relationship between steady state lead concentration and flow-rate under flushing conditions— Pipes B and C (soft water)

the experiment. Apart from the much higher lead levels, the most noticeable difference between these results and the results for pipe A was the much larger degree of scatter of the observations. However, it can be seen that the general pattern of the data conforms with that shown by the hard water pipe, particularly at high flow-rates (> 10 min^{-1}) where a definite upturn in lead concentrations was again observed in both cases. Fig. 2 also shows that flushing lead levels for pipe C were consistently lower than for pipe B. This is presumably because, under flushing conditions lead concentration depends on pipe length and pipe C is considerably shorter* than pipe B (Table 1, Sect. 2). It should be noted that the maximum flow-rate for pipe C was much greater than for pipes A and B due to higher mains pressure. Flow-rates above about 20 litres per minute are impossible to measure accurately by the method used in these experiments (Sect. 2.1) due to excessive splashing when collecting the water.

One possible explanation for the observed increase in total lead concentration at higher flow-rates is that particulate material is dislodged from deposits on the pipe's internal surface by the scouring action of turbulent flow. The results of filtration experiments designed to show how the proportion of non-filtrable lead material in tap-water varies with flow-rate (Tables 2 and 3, Section 3.1.1) support this hypothesis. It was shown, for both pipe locations, that the fraction of non-filtrable lead ranged from between 2 and 5% of the total lead at low-moderate flow-rates (0.5-7 litres per minute) to between 10 and 37% at high flow-rates (> 8 litres per minute).

Fig. 1 also shows that the increase in lead concentration at high flow-rates was considerably greater after the pipe had been left unused, compared with when a prolonged period of flushing was imposed before the start of the experiment. It seems likely that pipe deposits formed during periods of flushing would be more adherent than those formed during prolonged periods of stagnation, simply because the looser deposits would be washed away before they had time to build up appreciably. No work was done to determine the effect of prior water use at a tap on the

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* In addition, the internal diameter of pipe C is larger than pipe B, resulting in a smaller surface area exposed per unit volume of water.

non-filtrable lead fraction because it was not possible to impose realistic domestic water use patterns at the experimental locations.

The results of these experiments suggest that the optimum flow-rate range for taking tap-water samples under conditions of continuous flushing lies between about 3 and 7 litres per minute. Within this range the lead concentration approaches a minimum and is therefore less sensitive to flow-rate fluctuations.

Similar conclusions could also apply to the optimum flow-rate for taking samples after stagnation. However, although lead levels increased with increasing flow-rate for pipes A and B (Figures 4 and 5), the effect appeared to be of little practical importance within the flow-rate range investigated.

In practice, flow rates between 3 and 7 litres per minute are classed as 'moderate' flow-rates and would not be unrepresentative of typical domestic flow-rates, except perhaps for filling kettles where much higher flow-rates might be used.

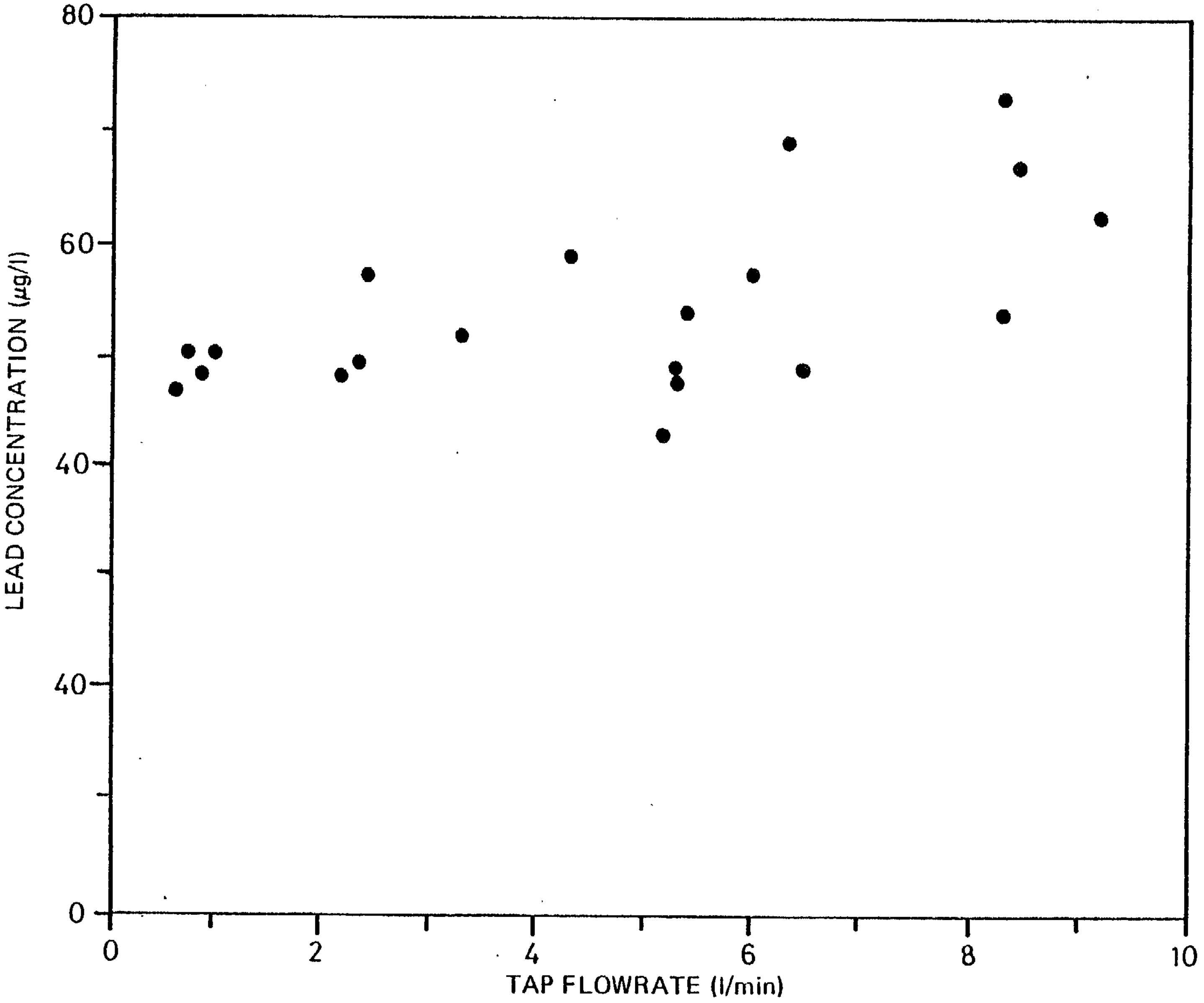

Fig. 4. To show the effect of sampling flow-rate on lead concentration at constant 5 min stagnation (Pipe A – hard water)

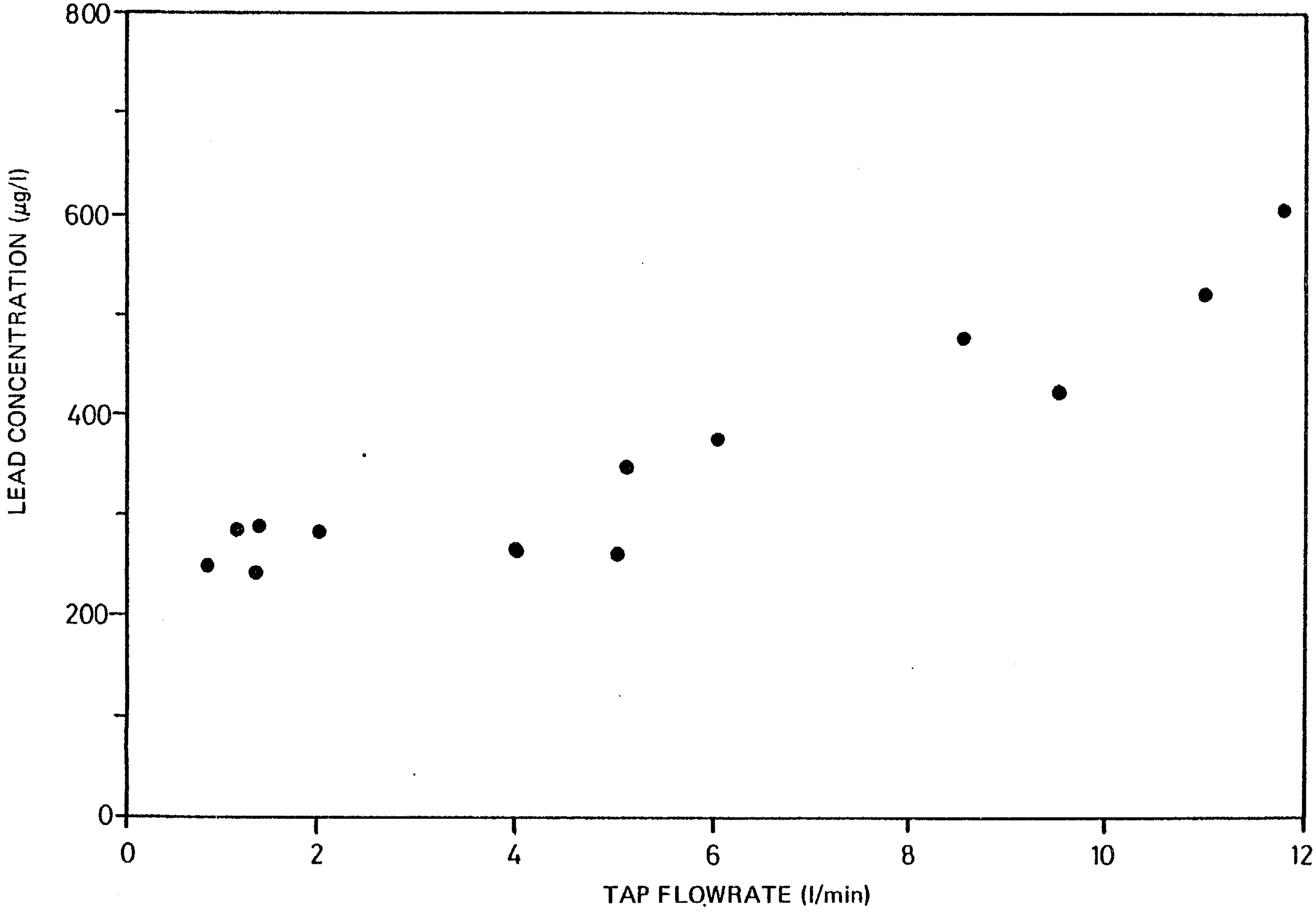

Fig. 5. To show the effect of sampling flow-rate on lead concentration after a constant 5 min stagnation (pipe B – soft water)

3.1.1. Filtration of tap-water samples

Tables 2 and 3 below show how the non-filtrable lead fraction varies with tap flow-rate. In this experiment, the non-filtrable lead fraction is defined as the proportion of the total lead in the sample which does not pass through a 0.4 micron pore size Nuclepore membrane filter.

Table 2 Pipe A (Hard Water)

Tap flow-rate (litres min^{-1})	Non-filtrable lead fraction as percentage of total lead in sample
0.45	1.8
0.8	2.0
1.2	2.1
3.1	2.5
3.3	2.3
4.9	5.2
5.2	4.6
6.3	3.5
6.6	10.0
8.2	11.8
8.9	37.3
9.2	25.0

Table 3 Pipe B (Soft Water)

Tap flow-rate (litres min^{-1})	Non-filtrable lead fraction as percentage of total lead in sample
0.6	3.4
0.8	2.6
1.0	3.9
4.7	4.3
11.9	24.4
>30	20.4
>30	20.3

3.2. EXPERIMENTS UNDER STAGNATION CONDITIONS - ALL PIPES

Assuming water quality remains constant, the stagnation time of water in the domestic lead service pipe and house plumbing is the most important factor controlling lead levels in drinking water. The stagnation curves for the three pipes used in this study (Fig. 3) all exhibit the same features: a rapid initial rise in concentration from the flushing base level in the first few hours, levelling off to an upper steady state level after about 10 to 20 hours. Similar experiments on old and new experimental lead pipes by Elzenga(9) at KIWA* in the Netherlands have produced very similar stagnation curves, which reach the maximum steady state level at around the same time.

Early attempts at representing the stagnation curves mathematically were based on some form of exponential decay expression. Three models relating lead concentration to stagnation time (T) were fitted:

(i) $$[Pb] = C - \alpha e^{-\beta T}$$

(ii) $$[Pb] = C - \alpha e^{-\beta\sqrt{T}}$$

(iii) $$[Pb] = F + \alpha \ (1 - e)^{-T/K\beta}$$

(Three further models were estimated using the dependent variable log [Pb] in order to improve the residual error structure).

No model was satisfactory for all sets of data. Model (ii) was an improvement on model (i), but both (ii) and (iii) suffered from the disadvantage that external estimates had to be supplied - or approximated iteratively - for the parameters C (the asymptotic upper limit), F (the 'base level' under flushing conditions) and K.

These difficulties were largely circumvented by a subsequent model which, in addition to being simpler mathematically, fitted all data sets

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* Keuringsinstituut voor Waterleidartikelen, Rijswijk, The Hague, Netherlands. (The Testing and Research Institute of the Netherlands Waterworks)

better than any of the previous models. The relationship is:

$$\log_e [Pb] = \alpha - \frac{\beta}{(T + \theta)}$$

The performance of this model is illustrated in Fig. 3 where it can be seen that it provides a good fit for each of the three sets of stagnation data. The parameter θ must be optimised by an iterative procedure, but experience in fitting the model to a wide range of stagnation data has shown that the fit is not very sensitive to changes in θ. The optimum θ value was usually between 20 and 100 minutes.

In all the stagnation experiments, a 'fully flushed' sample (ie, zero stagnation time) was taken before each stagnation period (Section 2.2). This raised the question of whether paired differences should be formed, basing the model on lead in excess of the flushing base level, or whether the zero stagnation time lead values should simply be treated as additional data points at the bottom end of the curve. The answer depends on the relative magnitudes of:-

(i) the true fluctuation in base level lead concentration from day to day,

(ii) the within-day variation in base level lead concentration,

(iii) the analytical error in determining lead concentration.

If there is little real variation in the flushed base level from day to day there is no advantage in forming paired differences - on the contrary, it will increase the experimental noise in the data. But if flushed concentrations are treated as independent observations when they do vary substantially from day to day, the data will follow a curve that shifts through time and so will be more scattered than had pairing been adopted.

Both approaches were tried on the data for the curves shown in Fig. 3. In all three cases, it was found that pairing the observations gave a worse fit than using the base level results separately. It can, therefore, be concluded that, under these particular experimental conditions, the flushed base level lead concentration is essentially stable for the duration of the experiment.

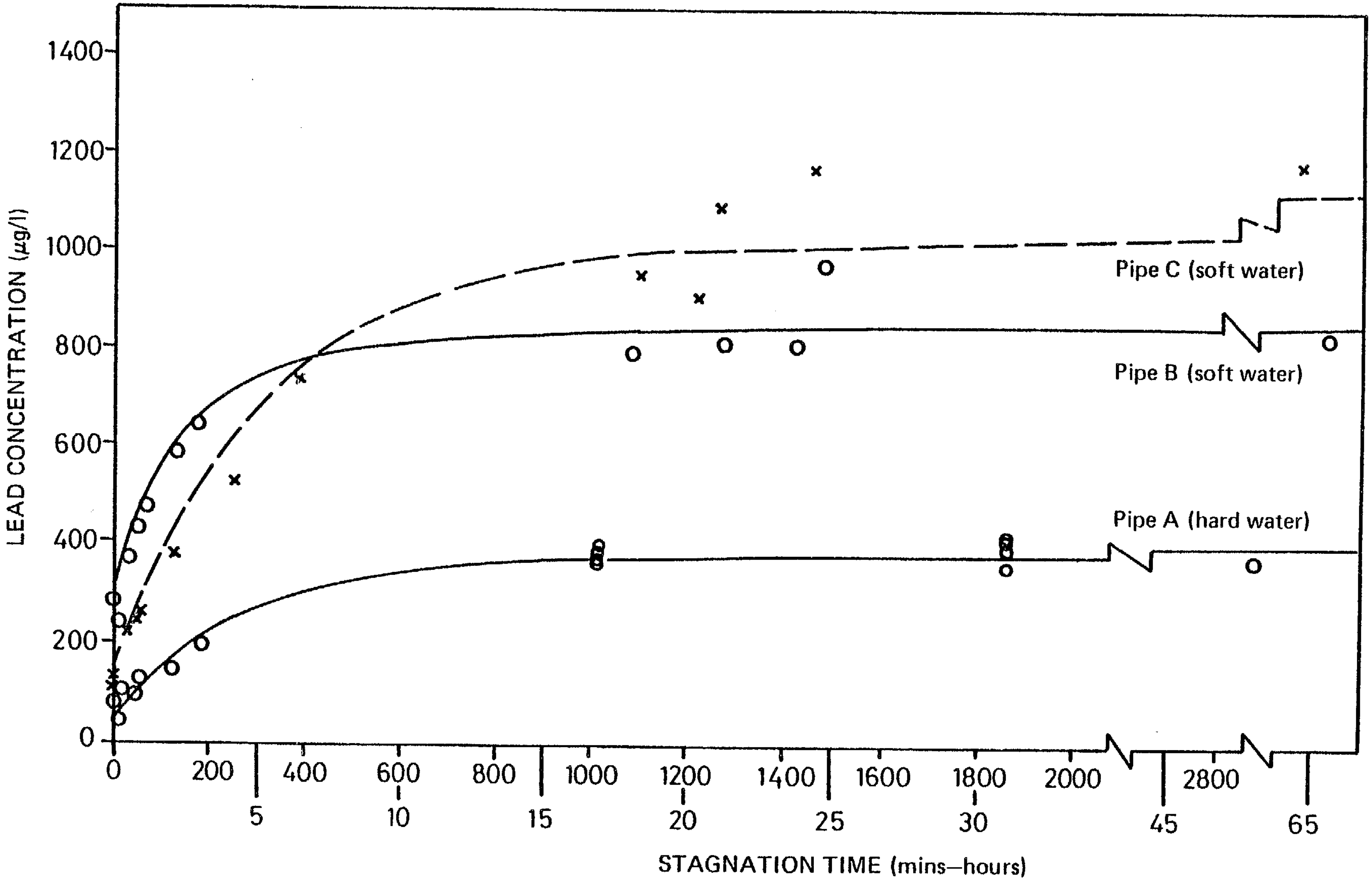


Fig. 3. Typical stagnation curves

From the practical viewpoint, these results suggest that it is not necessary to take a base level sample corresponding to each stagnation sample, provided sufficient replicates are taken over the experimental period to define the mean base level with a reasonable degree of precision.

3.2.1. Effect of sampling flow-rate on lead levels in stagnation samples.

The results from the experiment designed to assess the effect of sampling flow-rate after a constant stagnation period of 5 minutes are presented in the form of scatter diagrams for Pipe A (Fig. 4) and Pipe B (Fig. 5). In spite of the high degree of scatter in both sets of data, the lead concentration increases significantly with increasing flow-rate.

4. CONCLUSIONS

The work presented in this Appendix has shown that flow-rate and stagnation time both have an influence on lead levels in drinking water from consumers' taps. The influence of stagnation time greatly outweighs that of flow-rate.

Flow-rate

Under conditions of continuous flushing of the tap, lead concentrations decreased with increasing flow-rate up to about 3 litres per minute and then began to level off. Increasing the flow-rate above about 7 litres per minute usually produced an increase in lead concentrations, which was attributed to the scouring of particulate material from the pipe walls as a result of increased turbulence.

Experiments conducted to discover how the proportion of insoluble lead varied with flow-rate showed large increases in the non-filtrable (particulate) lead fraction with increasing flow-rate above about 7 litres per minute.

Under stagnation conditions, where the stagnation time of water in the pipe was held constant, increasing the flow-rate at which samples were taken from the tap increased lead concentrations in the samples. However, the increases were much less pronounced than under continuous flow conditions and little practical importance was attached to this effect.

Stagnation time

The effect of stagnation time of water in lead pipes was very marked. Lead concentrations increased rapidly in the first few hours of stagnation and levelled off somewhere between 10 and 20 hours. A simple mathematical model was developed which adequately described all the stagnation curves obtained experimentally.

REFERENCES

1. BAILEY, R.J. and RUSSELL, P.F. Trace Elements in Drinking Water. WRC Conference on Water Distribution Systems held at Keble College, Oxford, September 1978. Papers and proceedings published November 1978.

2. AINSWORTH, R.G., BAILEY, R.J., COMMINS, B.T., PACKHAM, R.F. and WILSON, A.L. Lead in Drinking Water. WRC Technical Report TR 43, 1977.

3. CRAUN, G.F. and McCABE, L.J. Problems Associated with Metals in Drinking Water. Journal AWWA, November 1975, 593-599.

4. CARTER, J.M. Lead in Drinking Water. Water, September 1977, 2-5.

5. DEPARTMENT OF THE ENVIRONMENT. Lead in Drinking Water: A Survey in Great Britain 1975-1976. HMSO Pollution Paper No. 12, 1977.

6. MOORE, M.R. Lead in Drinking Water in Soft Water Areas - Health Hazards. Science of the Total Environment, 1977, 7, 109-115.

7. RICHARDS, W.N., BRITTON, A. and COCHRANE, A. Reducing Plumbosolvency - the Effect of Added Lime on the Loch Katrine supply to Glasgow. Journal IWES, 1980, 34 (4), 315-333.

8. HUNT, D.T.E. Filtration of Water Samples for Trace Metal Determinations. WRC Technical Report TR 104, 1979.

9. ELZENGA, C.H.J. Proposal of the European Communities for a Directive Relating to the Quality of Water for Human Consumption. IWSA Congress on the Corrosion and Protection of Underground Pipelines held in Paris, September 1980.

APPENDIX 2 B

FACTORS AFFECTING COPPER LEVELS IN DRINKING WATER

CONTENTS

APPENDIX 2B - FACTORS AFFECTING COPPER LEVELS IN DRINKING WATER

Following the work on lead (reported in Appendix 2A) the influence of stagnation time and flow-rate on tap-water copper levels was studied.

1. STAGNATION TIME

Stagnation experiments have been completed on four *in-situ* aged copper pipes. Three pipes were in soft water areas and one was in a hard water area.

Two of the soft water areas were selected because it was known that the low pH values of the water gave rise to generally high tap-water copper levels (Ayr and Wigan). The third soft water area (Glasgow) was selected because recent pH correction with lime has raised the pH of a previously highly aggressive water. As a previous study(1) had shown that hard borehole waters tended to give rise to high copper levels in drinking water, a hard water area (Maidenhead) was also selected. A brief summary of the pipe characteristics and water qualities is given in Table 1.

Table 1. Details of *in situ* copper pipes and their respective waters.

	Ayr*	Glasgow*	Wigan*	Maidenhead
Diameter of bore (mm)	13	13	13	13
Approximate length (m)	9.0	8.0	9.5	9.8†
Calculated volume (litres)	1.2	1.1	1.3	1.6
Total hardness (mg $CaCO_3 l^{-1}$)	12	14	43	346
Alkalinity (mg $CaCO_3 l^{-1}$)	< 5	< 5	15	281
Total Organic Carbon (mg C l^{-1})	2.0	1.4	2.3	NA
pH	6.2	8.2	6.7	7.5

*Ayr, Glasgow and Wigan pipes were discussed in a previous CEC progress report(2).

†First 2m has a diameter of 19.5 mm.

All pipes had qualitatively similar stagnation curves. Mathematical functions were fitted to the data using a computer program (GENSTAT).

The forms of the curves tried are as follows:-

$$\log_e [Cu] = \alpha + (\beta/T + \theta) \qquad \text{(i)}$$

$$[Cu] = \alpha + (\beta/T + \theta) \qquad \text{(ii)}$$

where T is the stagnation time and α, β and θ are parameters estimated for fitting the curves to the data.

In all cases, function (i) fitted the data better than function (ii).

The calculated parameters are given in Table 2 and the fitted curves are shown in Figs. 1-4.

Table 2. Parameters of best line fit for copper pipes.

Parameter	Ayr	Glasgow	Wigan	Maidenhead
α	6.57	5.25	6.81	6.77
β	98.82	207.58	81.89	192.16
θ	35.75	80.45	32.74	51.02
No. of observations	32	42	39	63
% variance explained by model	99.2	95.5	97.4	97.6

2. FLOW-RATE

The relationship between copper concentration, under steady state, continuous flow conditions, and flow-rate was studied in Ayr, Wigan and Maidenhead. Fig. 5 shows the results for Ayr and Wigan; Fig. 6 shows the results for Maidenhead. The Wigan data were rather unexpected as the copper levels were independent of flow-rate, even at low flows (c. 1 litre $\min^{-1}$). In all cases there was no observable upturn in copper levels at high flow-rates as was observed for lead under some conditions (Appendix 2A, Section 3.1). Levels of copper when the Maidenhead pipe was flushed were very low (Fig. 6); only two observations, at low flow-rates, were above the analytical detection limit (15 $\mu g\ l^{-1}$).

3. CONCLUSIONS

Mathematical models similar to those developed for lead can be used to describe the relationship between the rate of build-up of copper concentration and stagnation time of water in copper service pipes.

Flow rate experiments gave results which were less conclusive than those obtained for lead. In one case, no dependence of copper levels on flow-rate was observed, even at low flow-rates and in another, copper levels when the pipes were flushed were below the analytical limit of detection.

Figure 1 - FITTED STAGNATION CURVE FOR AYR COPPER PIPE.

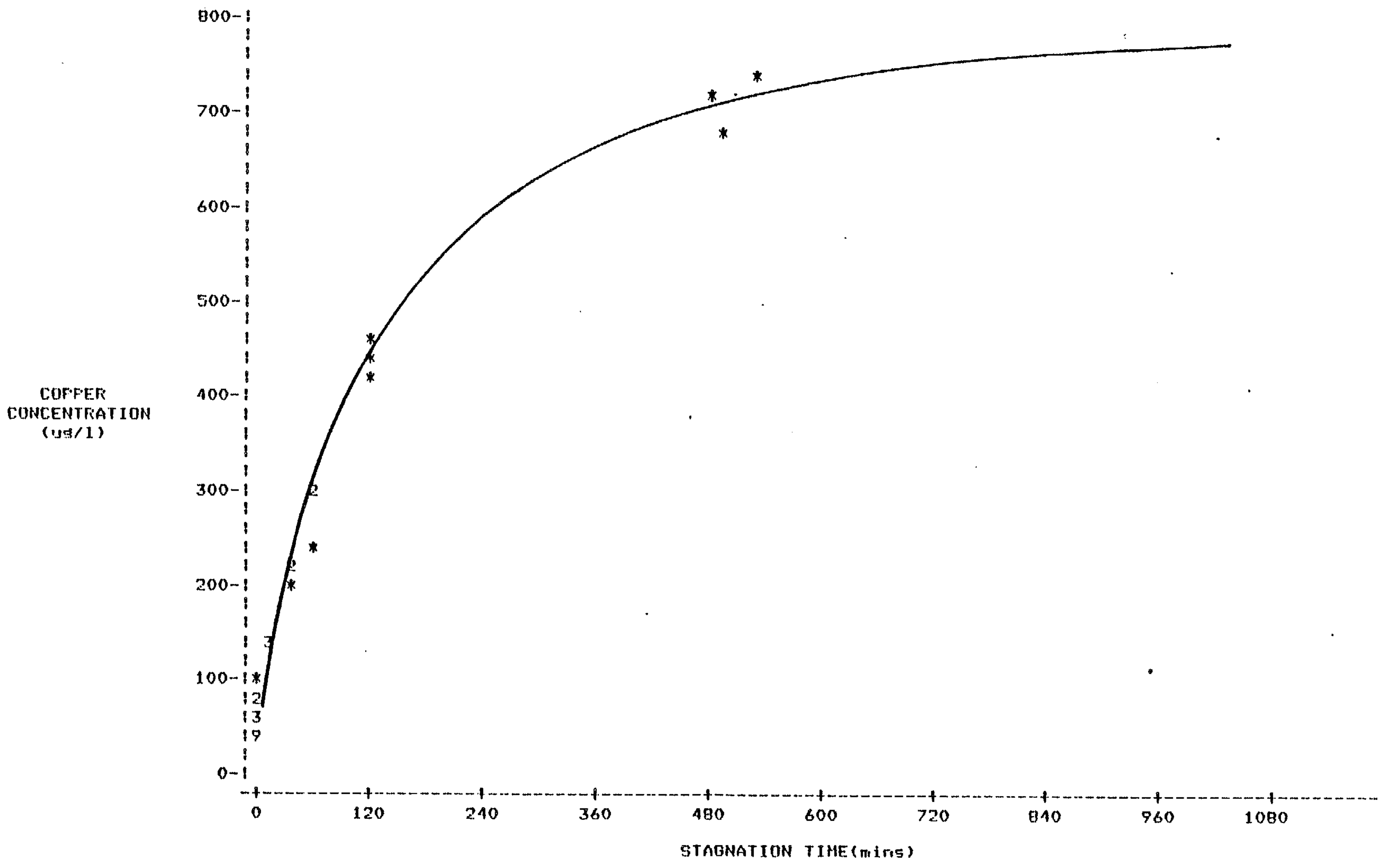


Figure 2 - FITTED STAGNATION CURVE FOR GLASGOW COPPER PIPE.

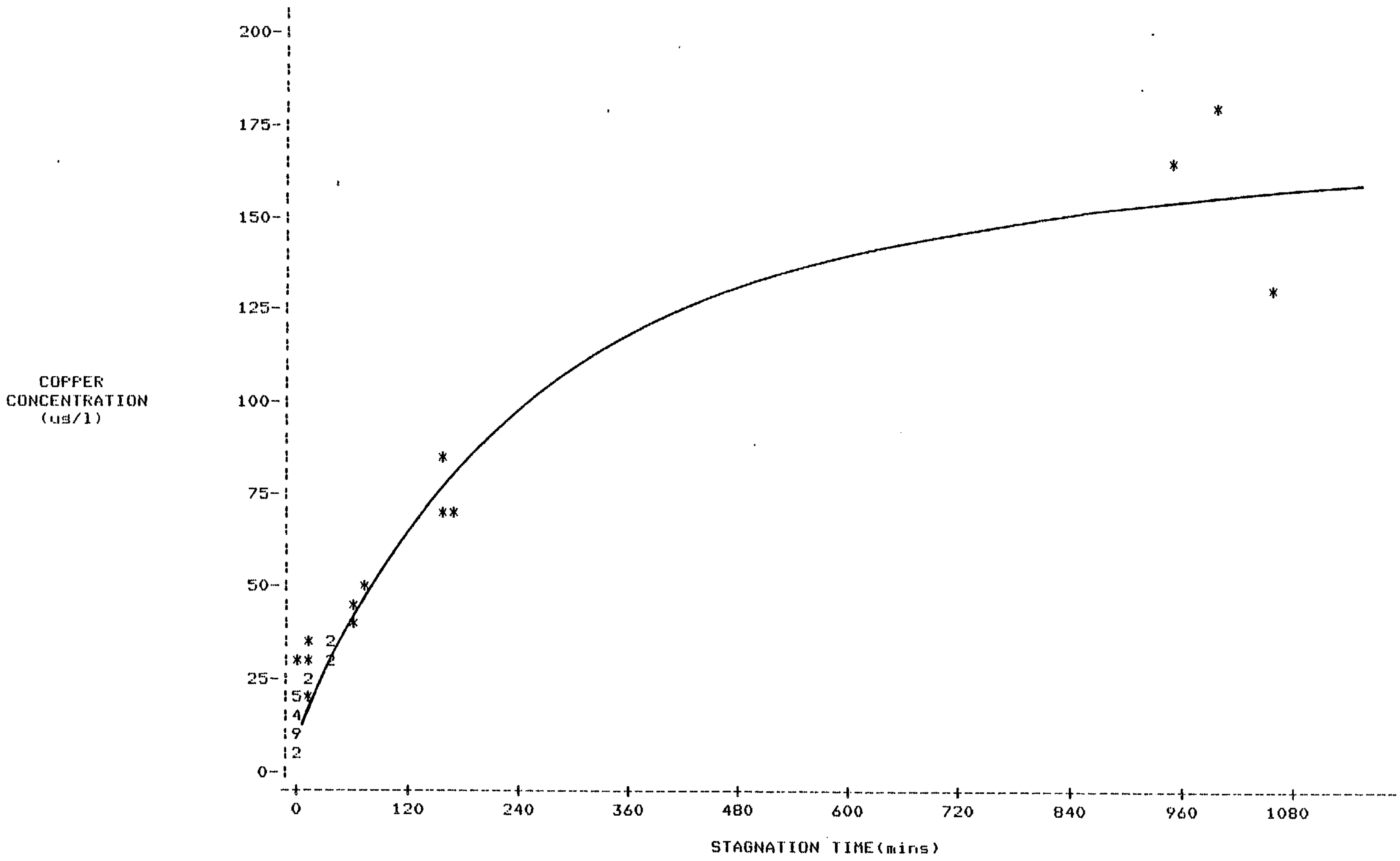


Figure 3 - FITTED STAGNATION CURVE FOR WIGAN COPPER PIPE.

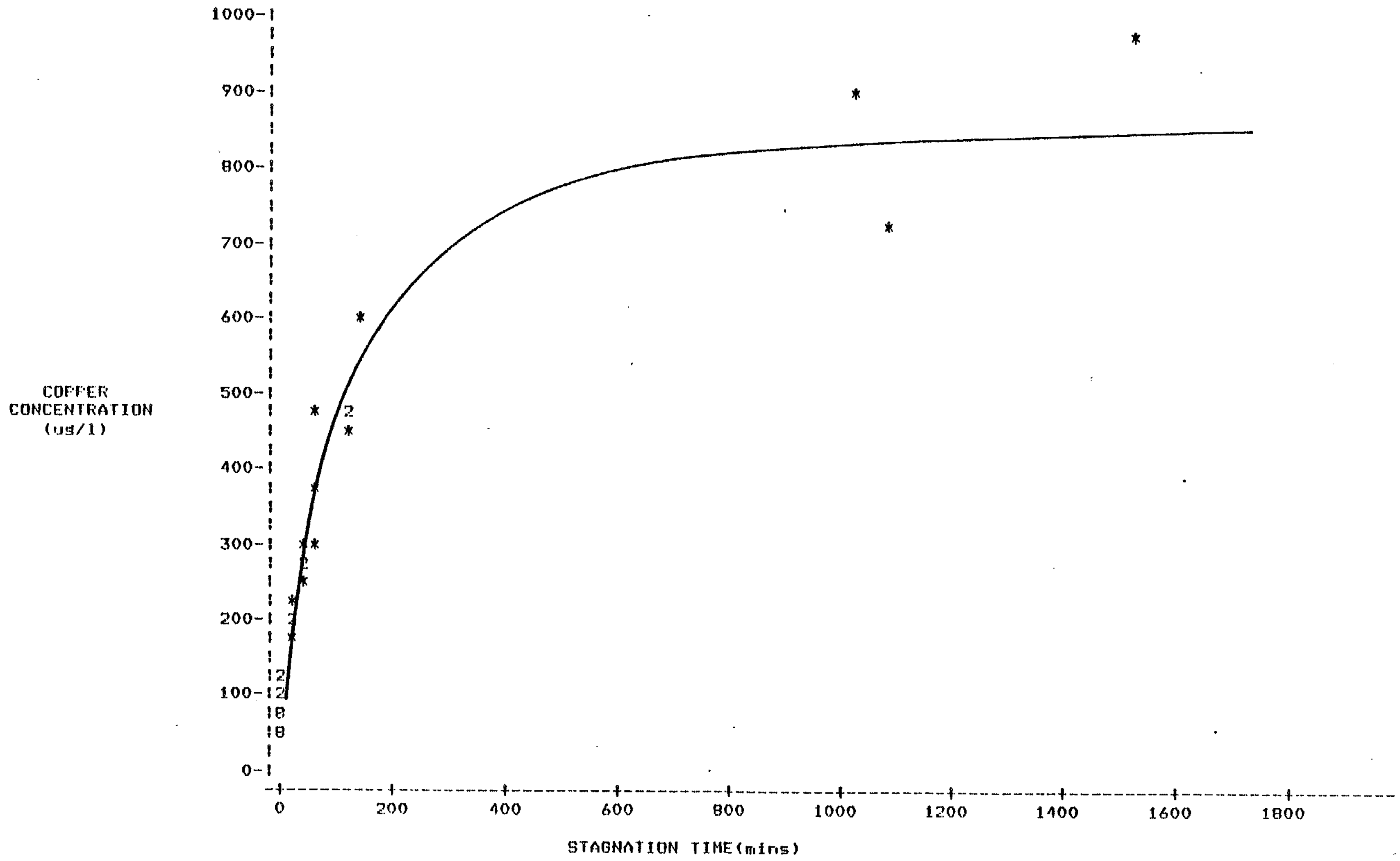
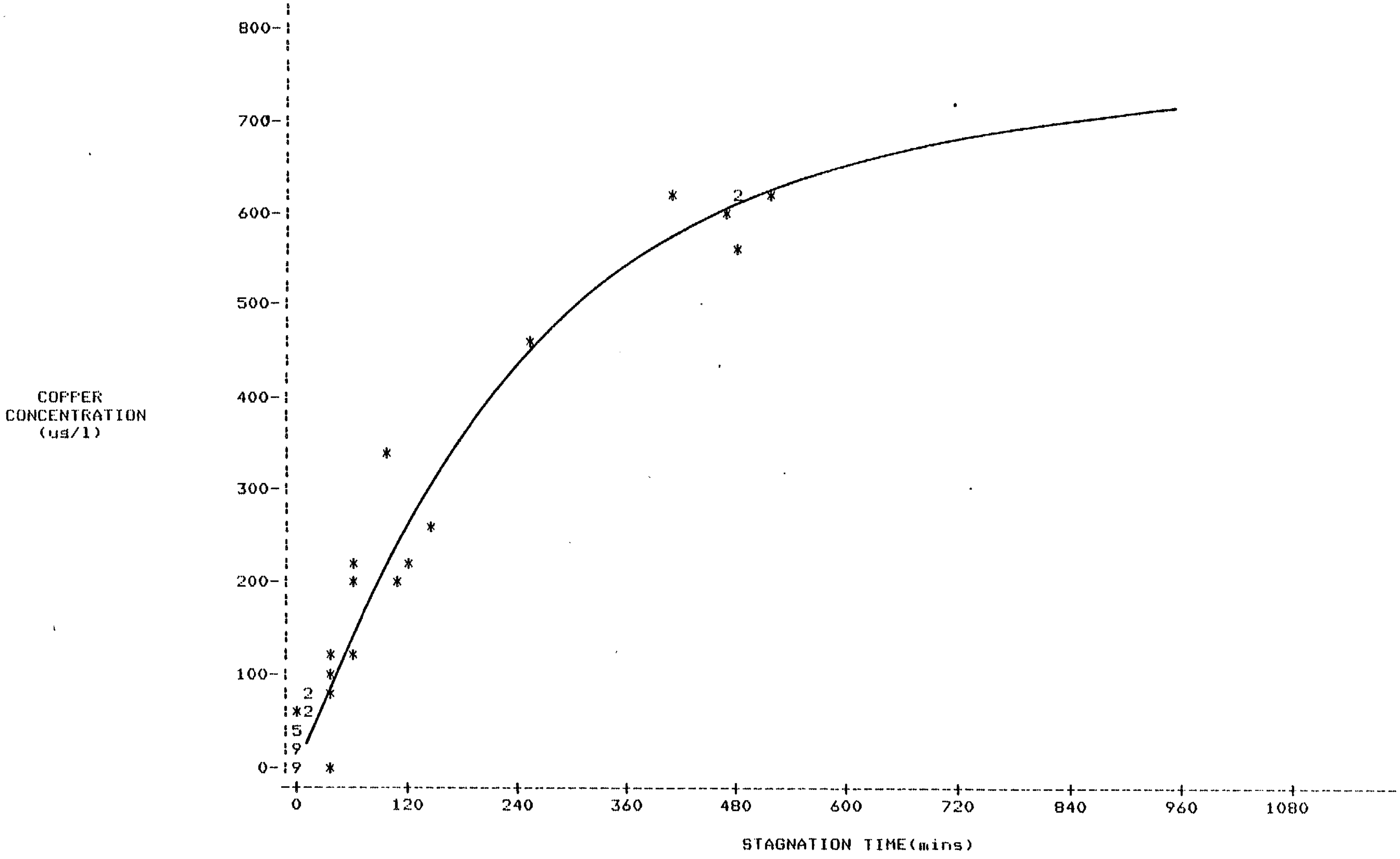


Figure 4 - FITTED STAGNATION CURVE FOR MAIDENHEAD COPPER PIPE.



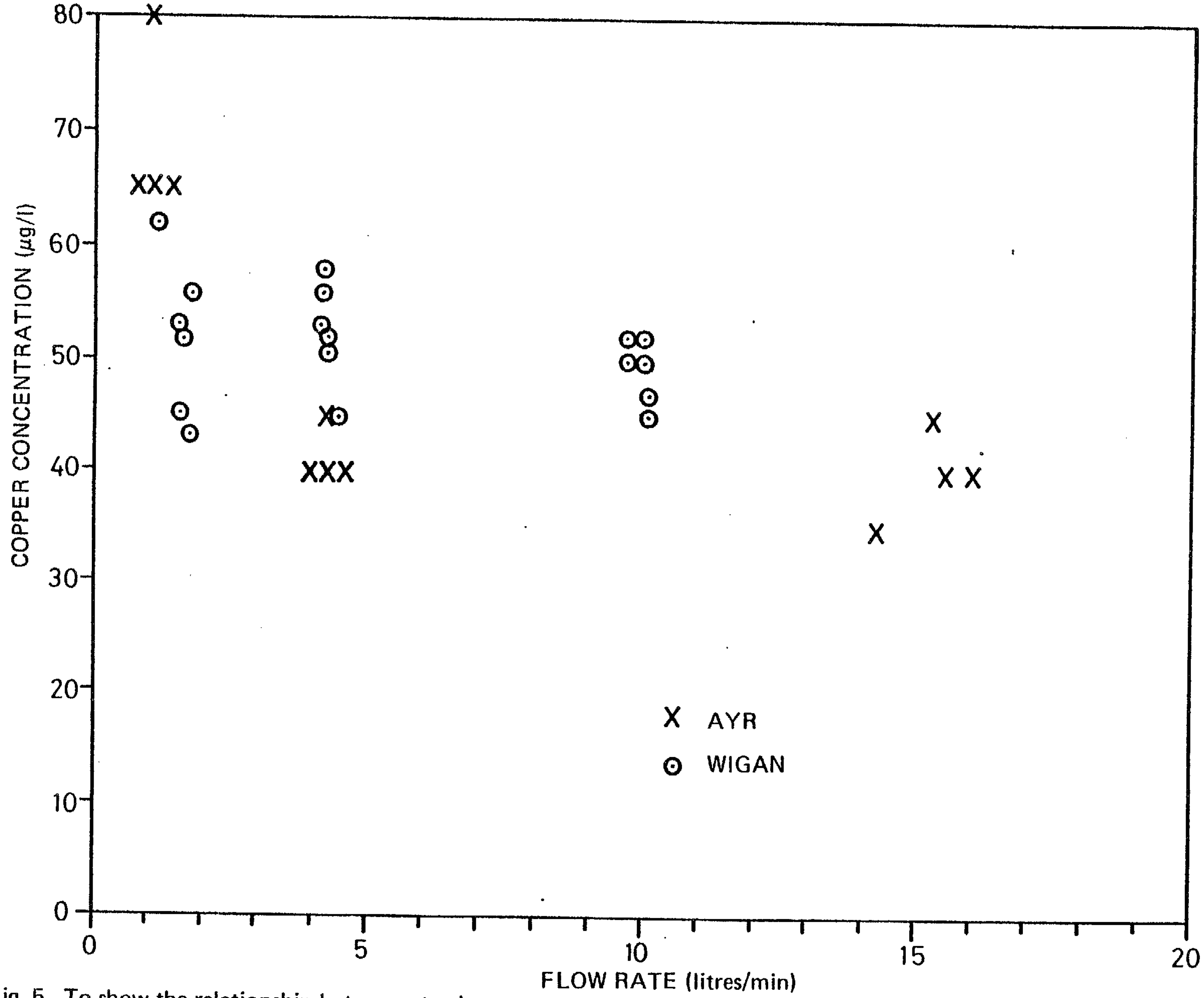


Fig. 5. To show the relationship between steady-state copper concentration and flow rate under flushing conditions (Ayr and Wigan pipes)

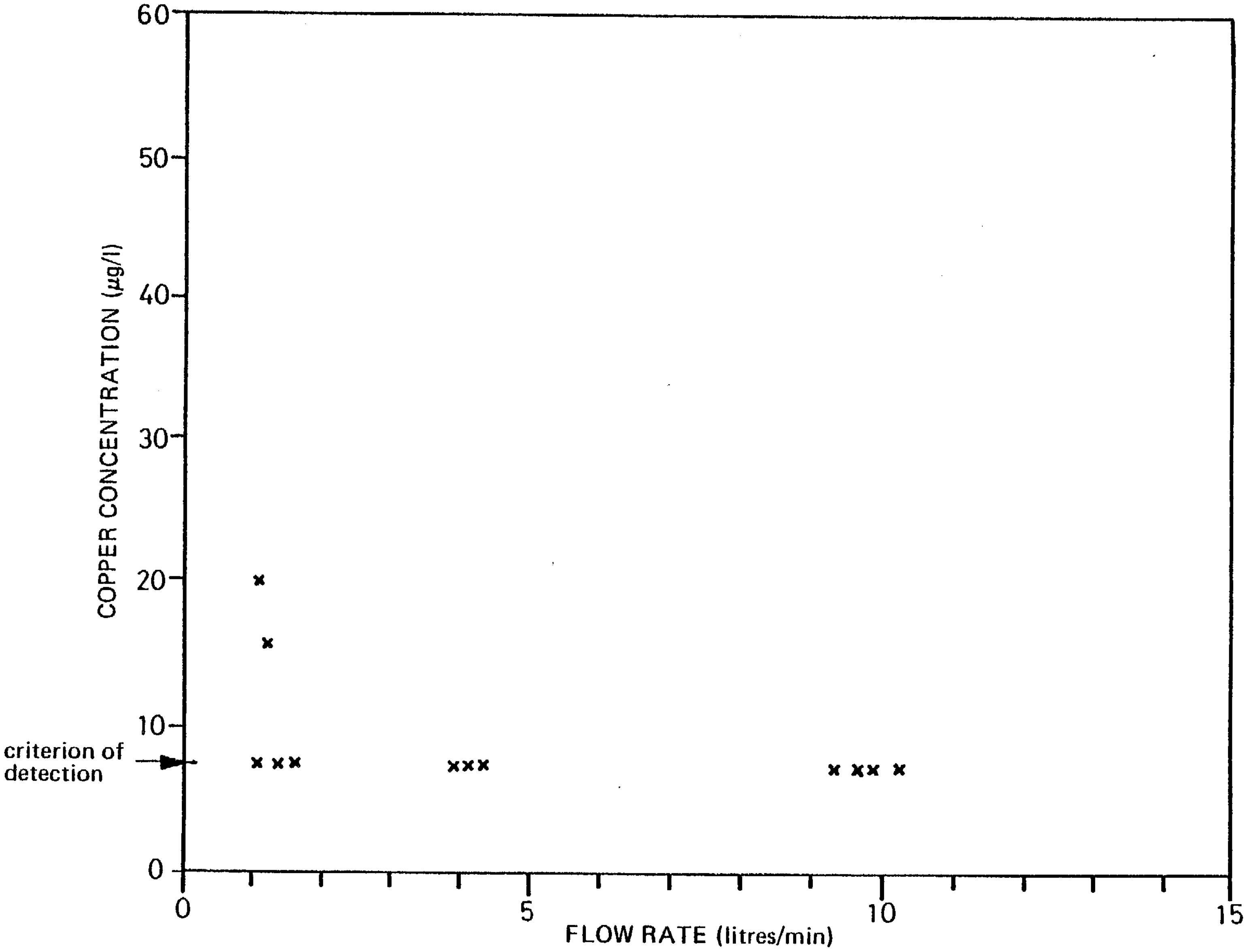


Fig. 6. To show the relationship between steady-state copper concentration and flow rate under flushing under flushing conditions (Maidenhead pipe)

REFERENCES

1. BAILEY, R.J. and RUSSELL, P.F. Trace Elements in Drinking Water. WRC Conference on Water Distribution Systems held at Keble College, Oxford, September 1978. Papers and proceedings published November 1978.

2. WATER RESEARCH CENTRE. Progress report to the CEC (July-December 1980).

APPENDIX 3

SAMPLING VARIATION AND PROPORTIONAL SAMPLING METHODS

CONTENTS

APPENDIX 3

SAMPLING VARIATION AND PROPORTIONAL SAMPLING METHODS

1. INTRODUCTION

Evaluation of an individual's exposure to lead from drinking water requires an accurate estimate of the concentration of lead in the water consumed. Lead concentration at the tap is very variable and is affected by factors such as the type and distribution of plumbing materials, the length of stagnation time of water in the pipe, type and quality of the water, temperature of the water and many other factors. It is important therefore to devise a sampling scheme that takes this variability into account and attempts to represent the actual exposure of a household to lead from drinking water. Two sampling schemes have been investigated and these are described below.

2. VARIABILITY OF FIRST-DRAW AND RANDOM-DAYTIME SAMPLES

The experimental procedure for estimating individual exposure of the Regional Heart Study respondents to trace elements from drinking water was to collect three discrete samples of tap water (first-draw, random-daytime and fully-flushed) from their kitchen taps. The results of the trace element analysis of these samples were then combined with information obtained from each respondent about the volume of each type of water and water-based drinks consumed per week. This measure of exposure will eventually be compared with CVD risk factors such as blood pressure to investigate possible associations between heart disease and constituents of tap water. It is therefore important to investigate the error surrounding this measure of exposure.

There are two main sources of error in the estimate of exposure. The first is the estimate of the intake of water and the proportions in which it is consumed (in terms of the three sample types), and the second is the variability of the lead concentration within each sample type.

It would be possible to obtain some indication of the first type of error by returning to a number of the respondents, asking the same questions about their drinking habits and comparing the two sets of answers (although this would not reveal a respondent's conscious or unconscious bias). However, in general, the majority of individuals have had regular drinking habits and therefore the estimate of volume is not thought to be a large source of error.

The second source of error, namely the variability of lead concentration within the sample type, can be investigated as a separate exercise. Such a study is now described.

2.1. METHOD

Approximately 16 random daytime and 16 first-draw samples were taken from each of 11 houses. The mean lead concentrations varied considerably between houses, and the within-house range increased with increasing mean lead concentration. Therefore, a log transformation was used to make the within-house variabilities more homogeneous. For each house, the mean of the logged values of all samples was obtained and each individual deviation about that mean calculated. Figures 1 and 2 show the lead values from 5 houses (first-draw and random-daytime results respectively) before and after the log transformation. The histograms of the logged concentrations show, in general:

i) less variation about the mean and

ii) the mean in each histogram is at approximately the same value.

This effectively put all the results on an equal footing so that they were directly comparable and could be pooled. Two histograms of the pooled residuals - one for first-draw and one for random-daytime results - were plotted and these are shown in Figs. 3 and 4 together with histograms of the original results. The 2.5 and 97.5 percentiles were read from the histograms of the logged results. The antilogs of these provided multipliers which, when applied to a particular lead value, would give an approximate 95% confidence interval for that house's geometric mean*. Alternatively, when applied to the geometric mean lead level for a house, the multipliers will give an approximate 95% central probability interval for individual sample values at that house. Examples for both these applications are given in the following section.

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* The geometric mean is the antilog of the mean of the logged values.

2.2. RESULTS

	2.5 percentile	97.5 percentile	antilog of 2.5 percentile	antilog of 97.5 percentile
First-draw	- 0.598	0.254	0.25	1.8
Random daytime	- 0.528	0.461	0.3	2.9

95% probability intervals for first-draw samples = (geometric mean value x 0.25) → (geometric mean value x 1.8

95% probability intervals for random-daytime samples = (geometric mean value x 0.3) → (geometric mean value x 2.9.

The standard deviations of the distributions of residuals ($\log_{10}$ values) for the first draw and random daytime samples (Figs. 3 and 4) were approximately 0.15 and 0.21 respectively. (These results would give an alternative method of estimating the multipliers if a log-normal form of distribution is assumed).

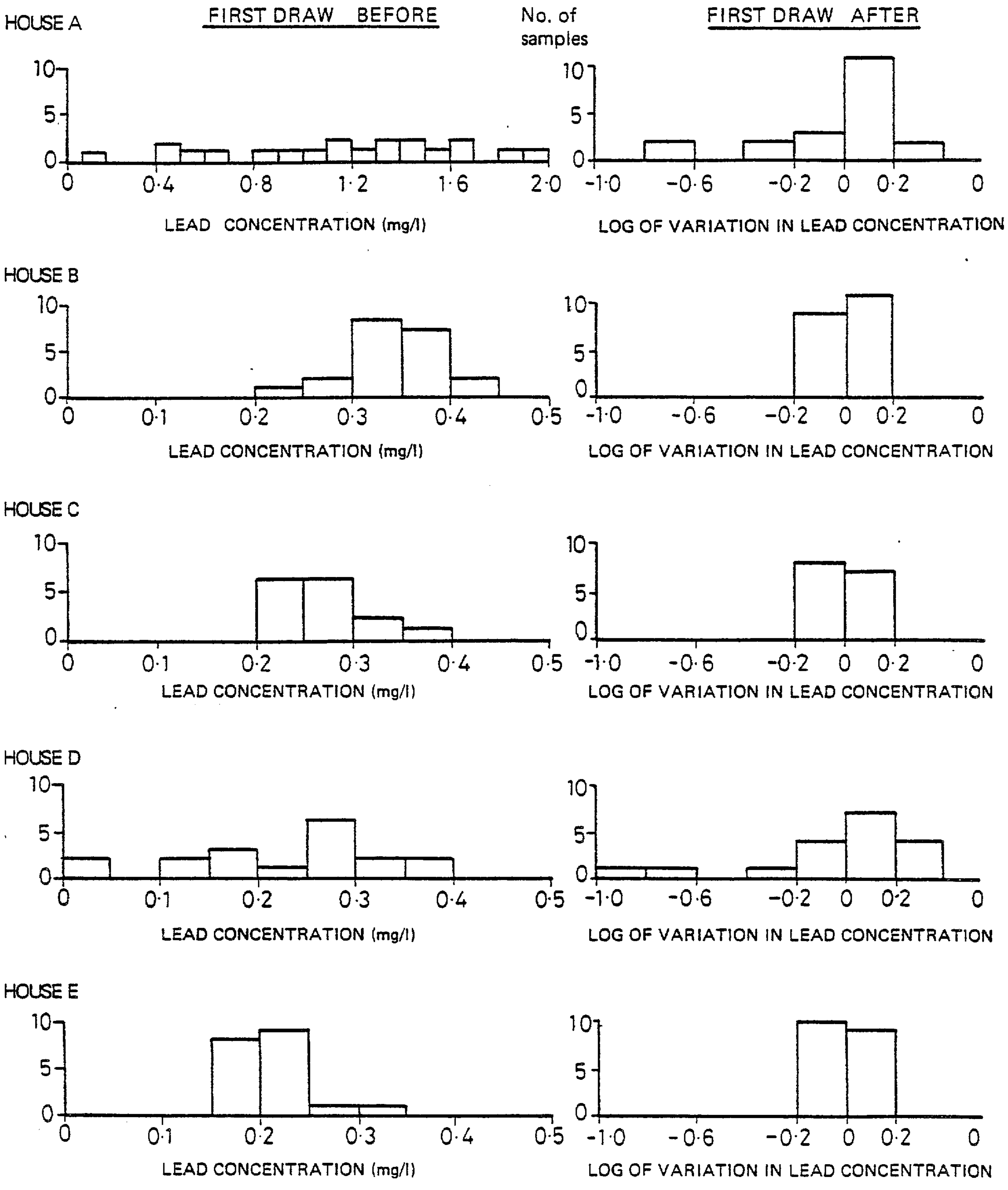

Fig. 1. Histograms of the first-draw lead concentrations from approximately 18 repeat samples at five houses before and after the log transformation

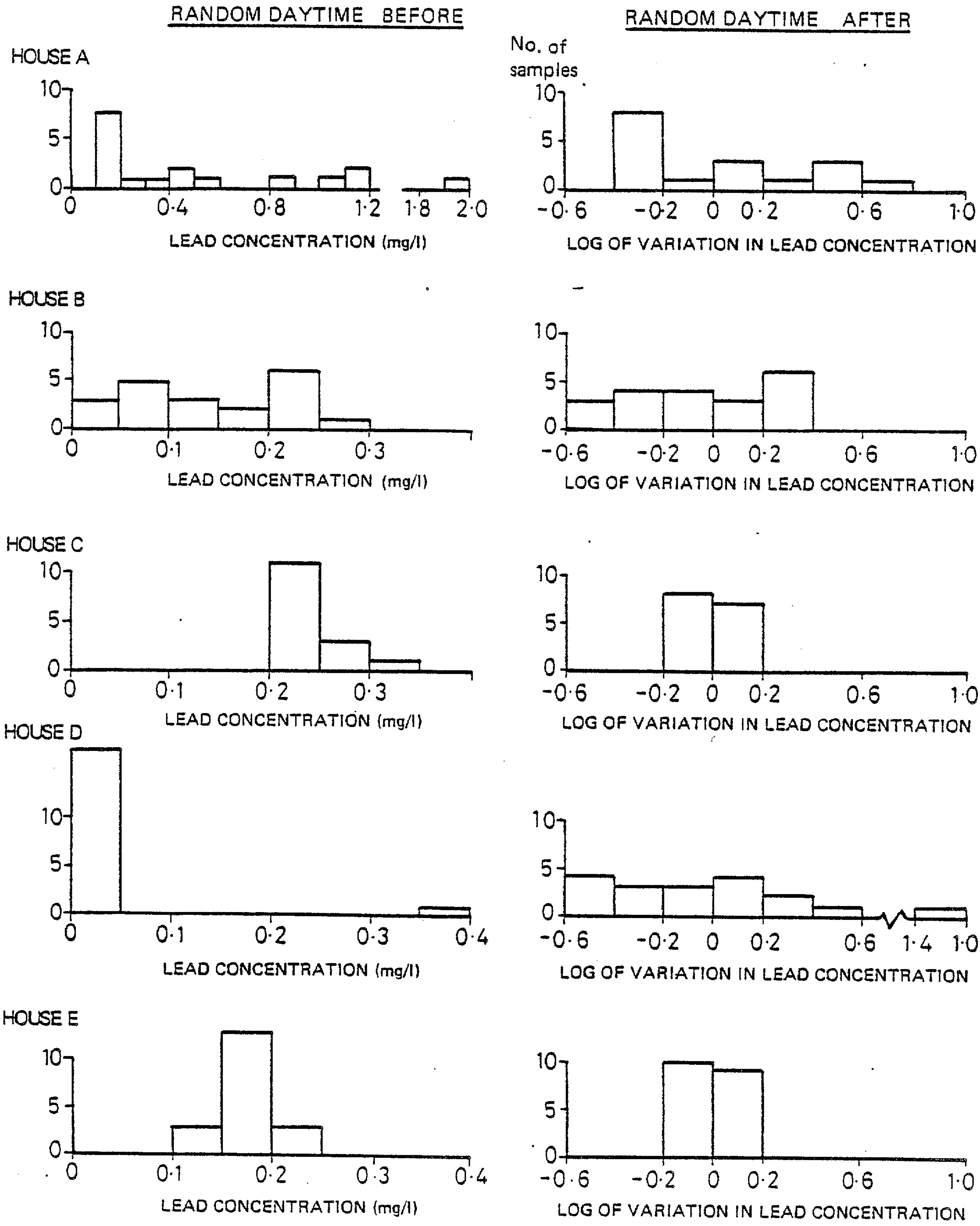

Fig. 2. Histograms of the random-daytime lead concentrations from approximately 18 repeat samples at 5 houses before and after the log transformation

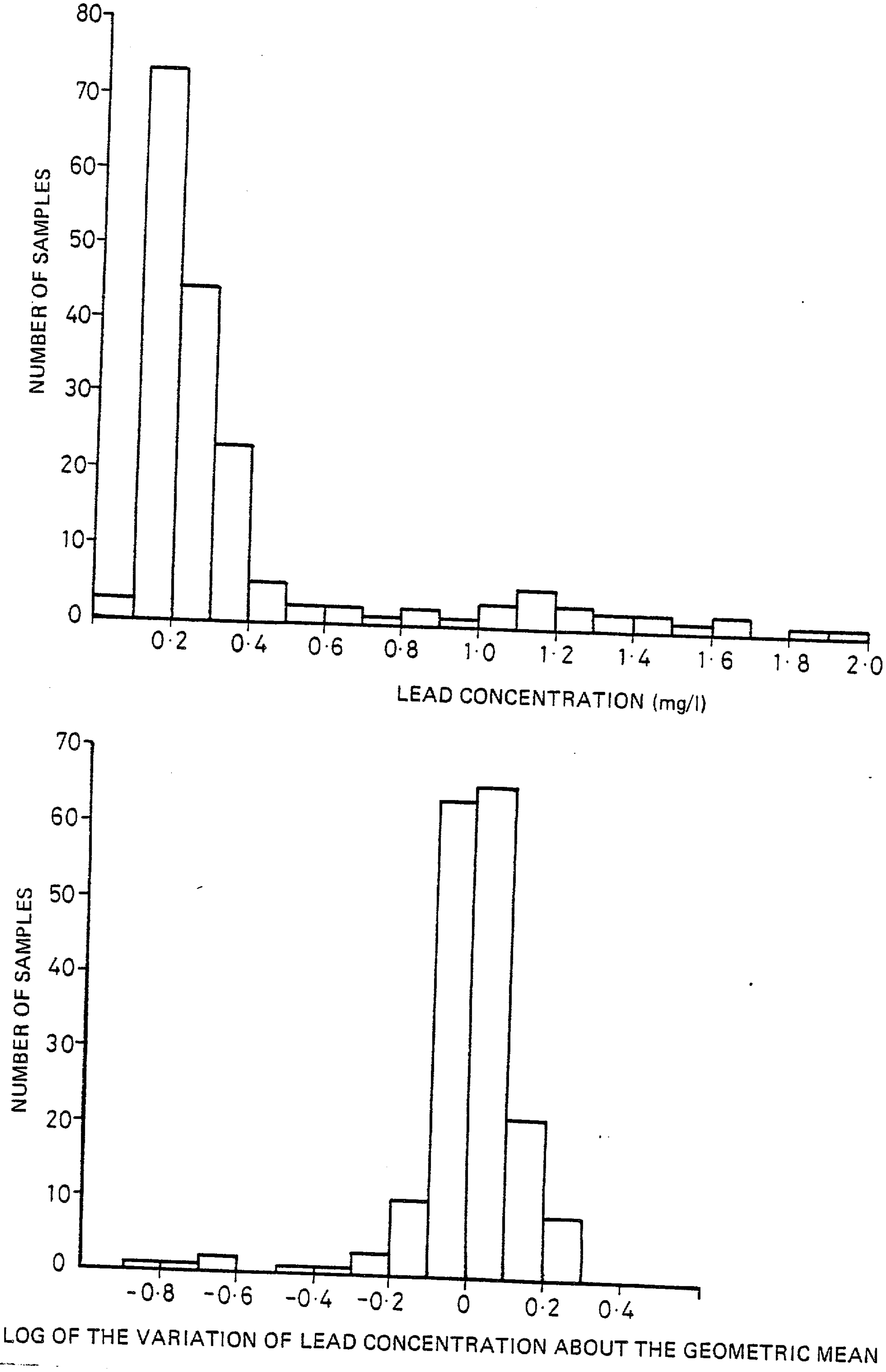

Fig. 3. Histograms of the first draw lead concentrations of approximately 16 samples from each of

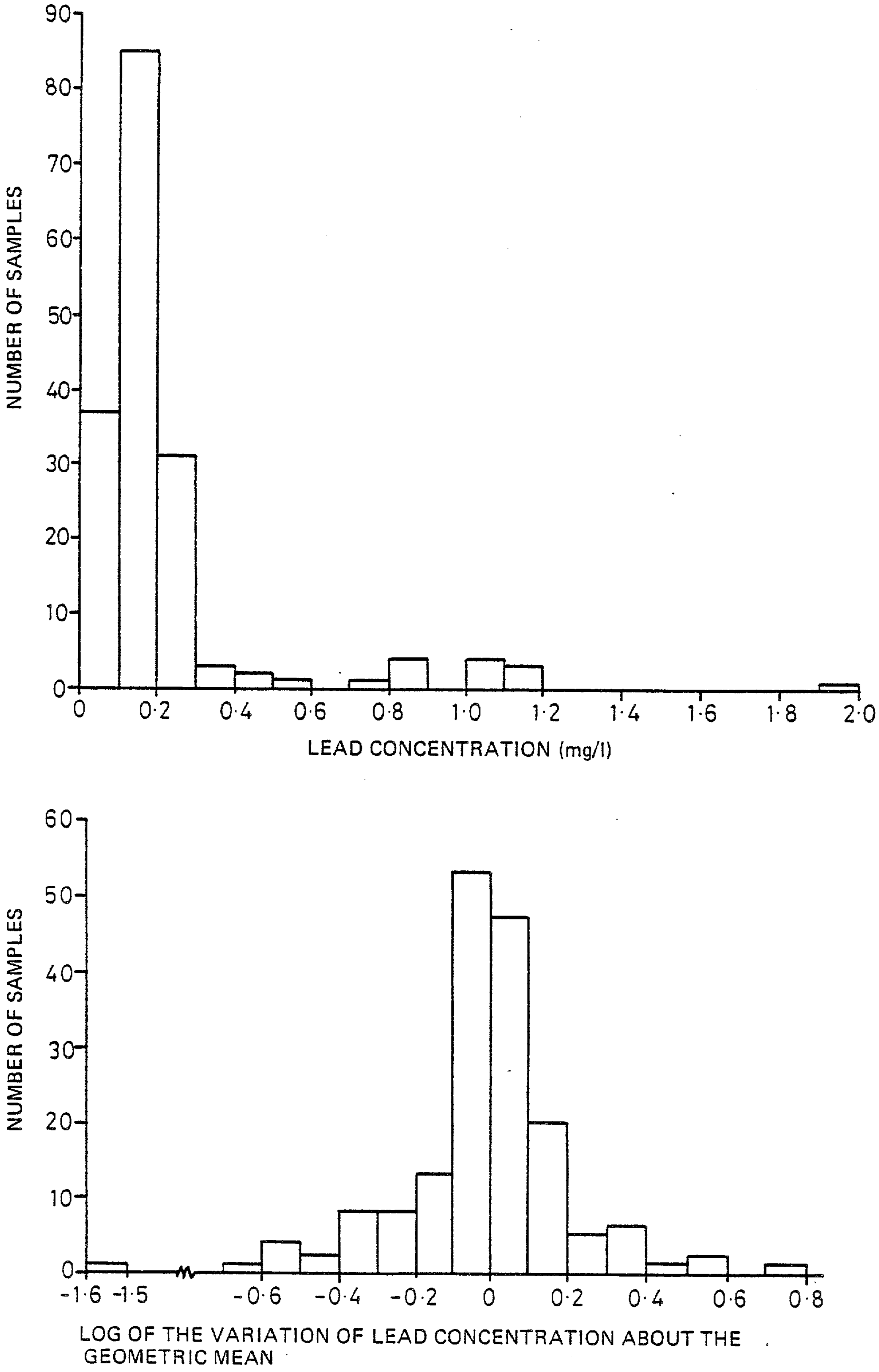

Fig. 4. Histograms of the random daytime lead concentrations of approximately 16 samples from each of the 11 houses before and after log transformation

Example 1

House F - 21 repeat first-draw samples - geometric mean = 0.16 mg l^{-1}.

95% probability interval = 0.16 x 0.25 ⟶ 0.16 x 1.8
= 0.04 mg l^{-1} ⟶ 0.29 mg l^{-1}

On average, 95% of values should fall between these limits (but not necessarily at one particular house, as is explained in the discussion). All twenty-one values are well within the limits in this instance.

Example 2

House A - 20 repeat first-draw samples - geometric mean = 0.87 mg l^{-1}.

95% probability interval = 0.87 x 0.25 ⟶ 0.87 x 1.8
= 0.22 mg l^{-1} ⟶ 1.57 mg l^{-1}

In this case, four values are outside the limits, compared with the expected value of one.

House A - 18 repeat random-daytime samples - geometric mean = 0.33.

95% probability interval = 0.33 x 0.3 ⟶ 0.33 x 2.9
= 0.1 mg l^{-1} ⟶ 0.96 mg l^{-1}

Again, four values are outside the limits, compared with the expected value of one.

Example 3

House M - (samples collected after main batch and therefore not included in original calculation)

Pick a first-draw value at random:- 0.3 mg l^{-1}

The geometric mean will be somewhere between

0.3 x 0.25 ⟶ 0.3 x 1.8

i.e. 0.075 mg l^{-1} ⟶ 0.54 mg l^{-1}, with 95% confidence.

The geometric mean of 19 first-draw samples at this house is 0.14 mg l^{-1}, which is within the confidence interval.

2.3. DISCUSSION

The method that has been used to calculate the multipliers is designed to apply to a range of lead concentrations from areas of different water quality. The data used in the calculations was from eleven houses with different variability of lead levels in order to achieve this aim. However, the houses are not all equally variable even after the log transformation. The 95% multipliers are therefore valid only for the town as a whole (assuming that the eleven houses are representative). In particular, the multipliers cannot be regarded as applying to any individual house. This is borne out in practice, and shown in the examples.

2.4. CONCLUSIONS

When trying to assess individual exposure to lead from drinking water by taking three discrete samples some error will be introduced because the concentration of lead at the tap is very variable and unpredictable. The calculations described above have shown that it is possible to quantify the error which is introduced. However, (1) due to the large between-house variability the confidence intervals should not be applied to an individual house, but only a group of houses and (2) the error bands are so wide that the result obtained from taking either one first-draw or one random sample from a house cannot be relied upon to give an accurate measure of population exposure.

3. PROPORTIONAL SAMPLING IN HOUSES - PRELIMINARY EXPERIMENTS

In the previous section a method of assessing population exposure using three sample types was described. In theory, a more reliable method of monitoring exposure would be to take a small proportion of the water actually consumed by a family. This can be achieved by attaching a proportional sampler to the kitchen tap in a number of households. The sampler proportions 5% of the water actually consumed by the householders and this can be collected in a sample bottle over a certain period of time (a week for example). There is a tap on the sampler which can be turned to the 'on' position every time water is used for drinking purposes and this allows a proportion of 5% of the total flow to enter a sample bottle.

Such a sampler has been designed in the Netherlands[*] and tested at the WRC to ensure that a) 5% of lead is proportioned and b) lead is not absorbed or leached by the sampler or tubing.

The purpose of this section is to describe two types of preliminary experiments that have been carried out to test a) whether households accept the sampler and use it correctly and b) whether the measure of exposure obtained using the proportional sampler is related to that obtained using the discrete sample approach.

3.1. METHOD

Initially samplers were installed in three households without lead plumbing for a period of one week. This test ensured that the sampler was correctly used, and the tap on the sampler turned on only when a drink was being taken. The household members were asked to complete a diary (shown in Fig. 5) noting each time a kettle was filled or a drink taken, the volume drawn, and the volume consumed. At the end of the week the volume collected in the sample bottle was compared with the figures from the diary for a) the volume drawn and b) the volume consumed.

The second set of experiments involved installing samplers in houses with lead plumbing for one week. Series of flushed, first-draw and random-daytime samples were also collected both prior to the installation of the sampler and after its removal. The exposure to lead from drinking water was then calculated in two ways:

a) using the results from the analysis of the discrete samples together with the information in the diaries about the volume of water consumed[+] by the family and

b) from the analysis of the proportional sample.

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[*] The proportional sampler was designed by B J A Haring of the Netherlands National Institute for Drinking Water Supply.

[+] Each household member also filled in a short questionnaire (shown in Fig.6) which yielded information on the proportion of first-draw, random daytime and fully-flushed water consumed.

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CODE

T = Tea W = Water
C = Coffee
H = Other hot drink
O = Other cold drink
M = Mug C = Cup

SURNAME ________________

DAY ________________

DATE ________________

Quantity drawn (pints)	Drawn into:- (K=kettle D=directly into drinking vessel)	Type of Beverage (Code)	Made by:- (Code letter)	Number of cups or mugs per person						
				A	B	C	D	E	F	V

Fig. 5. Dairy to be completed by householders using a proportional sampler

FAMILY DETAILS

1.

Code Letter	Name	Position in family (Father, etc.)	Sex M/F	Year of Birth (if under 21)
A				
B				
C				
D				
E				
F				

Questions 2 and 3 are to be completed by each individual.

2. Do you ever take or make a drink at home for yourself or any member of the family?

Please could each member of the family tick the appropriate box.

	A	B	C	D	E	F
Yes						
No						

3. When preparing a water-based beverage do you run the tap first or take the water straight from the tap?

Insert a 'D' for each occasion water is drawn directly from the tap, and an 'R' when some water is allowed to run to waste prior to taking water for drinking purposes. If you have no set pattern or do not prepare certain types of beverage, insert an X in the appropriate box.

Occasion of drawing water	A	B	C	D	E	F
Glass of water						
Filling the kettle						
Preparing baby food						
Other						

Fig. 6. Questionnaire to ascertain whether first-draw, random-daytime or flushed water is used in a house

3.2. RESULTS

Table 1 shows the results of the actual volume collected in the proportional sampler bottle compared with the volumes recorded in the diaries of the amount consumed and drawn.

Table 2 gives the results of the comparisons of the two estimates of exposure to lead from drinking water from the houses with lead plumbing.

3.3. DISCUSSION

Table 1 shows that, in general, the volume collected by the proportional sampler is higher than the diary figures for the volume of water drawn and that consumed. It is, however, far closer to the 'quantity drawn' than the consumed volume. This could be due to a number of reasons. For example, when tea-pots, coffe-pots or jugs are filled not all the liquid is necessarily consumed. Another reason could be that certain individuals may empty out the water remaining in the kettle, before refilling it. In all these cases water has been sampled by the proportional sampler, and not actually consumed. This, however, should not introduce a very large error into the estimate of exposure as the concentration of lead in the water that has been sampled is the same as that which has been consumed.

Table 1. Volume collected in the proportional sampler bottle (per week) compared with volumes recorded in the diaries of the amount consumed and drawn. (All volumes are in litres).

Location	Volume in proportional sample	Volume calculated from 'Quantity drawn'*	Volume calculated from 'Quantity consumed'*	% Error Quantity drawn / Prop.sample volume	% Error Quantity consumed / Prop.sample volume
Newmarket	1.0	1.3	1.1	130%	110%
Lowestoft	3.7	3.5	1.5	95%	40%
Bradford	1.4	1.1	1.1	79%	79%
Glasgow 1	1.5	1.4	1.1	93%	73%
Glasgow 2	3.5	2.2	1.2	63%	34%
Reading 1	1.6	1.7	0.35	106%	22%
Reading 2	0.7	0.7	0.5	100%	71%
Reading 3	1.9	0.9	0.8	47%	42%
MEAN	1.91	1.6	0.96	89%	59%

* Figures in these columns refer to 5% of the volume consumed by the family during one week.

Table 2. Comparison between exposure to lead from drinking water measured by the proportional sampler and that predicted from a knowledge of drinking habits and discrete sample concentrations. (All concentrations are in µg l^{-1}).

Location	Lead concentration in the proportional sample	Mean Random	Mean Flushed	Mean First draw	Predicted value	% error $\frac{\text{Predicted lead conc}}{\text{Proportional sample lead conc.}}$
Newmarket	11	6.5	4.5	11.5	6.1	55%
Lowestoft	56	30	27	82	30	53%
Bradford	21	26	10	73	23	109%
Glasgow 1	104	120	61	177	117	112 %
Glasgow 2	76	33	19	69	19	25%
						MEAN 71%

There is another possible explanation for the discrepancy in volume and this could have a more serious effect on the overall estimate of exposure. Certain individuals always flush the tap before taking water for drinking. If the sampler tap is on while the tap is flusing, as well as while the drinking water is being drawn, then the volume of water collected will be greater than that drawn. Moreover, the concentration of lead in the sample bottle could be greater than that consumed, particularly if this flushing occurs first thing in the morning or after water has not been used for some time in the house.

Table 2 shows that the concentration of lead in the proportional sample is considerably higher than the predicted value. This agrees with the explanation above regarding the sampling of water that is flushed away before a drink is taken.

The results from one household emerge as being particularly inaccurate (Glasgow 2). The householders always flush the tap before taking water for drinking purposes, therefore their predicted exposure corresponds to the mean 'flushed' value. However, the proportional sample lead concentration is higher than the mean first draw level.

This could be explained by the plumbing layout at the house. The kitchen is on the first floor. There is a 30 ft length of copper pipe running from the tap to the service connection, followed by about 50 ft of lead. It is possible that:

a) The first draw lead concentration is not a true reflection of the highest lead level from that house as the first litre which is drawn for the sample will have been stagnant in the copper piping.

b) When the householders flush the tap before taking water for drinking, they flush out the water that has been standing in the copper and drink the water that has been standing in the lead pipe.

3.4. CONCLUSIONS

The objectives of these experiments were to investigate

a) whether householders accepted the proportional sampler

b) if they used it correctly

c) whether the measure of exposure obtained using the proportional sampler was related to that obtained using the discrere sample approach.

All eight households that were asked to use the sampler did so to the best of their ability for a whole week - none abandoned the task. However, the volume of tap water consumed did not tally with the volume of water in the sample bottle. The sampler samples more water than is actually consumed and this is undoubtedly due to some kind of incorrect use of the sampler (for example, leaving the sampler tap open while flushing water to waste or while taking water for purposes other than drinking).

From the limited number of experiments and the large margin of human error it is impossible to say whether there is a relationship between the value of exposure obtained by the use of the proportional sampler and that calculated from drinking habits and discrete sample analyses. The lead levels are, in general, higher in the proportional sample than the predicted values, however, no definite relationship has emerged.

More experiments of this type are required in order to evaluate the sampler as a method of assessing exposure to lead. However, the main difficulty with this particular sampler is its dependence on human cooperation. An alternative solution to the problem could be to redesign the sampler, possibly to fit into the household plumbing system, in an attempt to minimise human error.

It is therefore suggested that future work should entail experimenting with the present design at the same time as investigating an improved design to minimise human error.

APPENDIX 4

RELATING TAP-WATER LEAD TO WATER USE PATTERNS

CONTENTS

APPENDIX 4 - RELATING TAP-WATER LEAD TO WATER USE PATTERNS

1. INTRODUCTION

It is recognised that variations in contact time of water with lead service pipes and plumbing, resulting from normal daily patterns of water use, give rise to a high degree of *within-house* variation in tap-water lead levels[1-5]. Although this variation is mainly due to the increase in lead concentration with increasing contact time in the pipes, other factors such as the flow-rate at which water is drawn from the tap, the rate of opening the tap and the volume of water drawn in relation to the length of pipe also contribute to the overall variation in lead levels. In addition, factors such as differing patterns of water use and varying lengths and type of lead pipe are likely to give rise to considerable *between-house* variation in water lead levels, even if the water quality remains constant.

The high variability of both within-house and between-house lead concentrations in drinking water means that very large numbers of samples are required to estimate typical lead concentrations in drinking water with reasonable precision. In practice, there are usually severe restrictions on the number of samples that can be taken, due to the high costs of both sampling and analysis.

One criterion for designing a survey to estimate exposure to lead in drinking water is that the samples should be representative of the water actually consumed. A recent survey of individual and household drinking water consumption patterns in Great Britain[6] showed that most of the tap-water drunk is drawn at random times throughout the day. By comparison the amount of water drunk that has been standing in the pipes overnight is extremely small.

Random daytime samples, as defined below* are therefore usually considered to be the most representative of consumed water and because they are quickly and easily collected are widely used to define typical or average exposure levels to lead in drinking water. However the high degree of within-house variation inherent in this type of sample means that an unacceptably large number of replicates is required to define mean drinking water lead

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* A sampler, calling at the houses, takes single samples from the kitchen cold taps at random times during the working day. No water is flushed from the tap beforehand. The stagnation time of water in the pipes before sample collection is not normally known.

levels at a particular house with reasonable precision. For example, six random daytime samples giving a household average of 60 μg l^{-1} would give a confidence interval (95%) around the true household mean as wide as 40 to 90 μg l^{-1}.

In cases where it is required to define maximum lead exposure levels, *first draw samples** are much more representative of water containing high lead concentrations than random daytime samples. In addition, the within-house variation in lead concentrations is expected to be less for first draw samples because of the lower variation in stagnation times compared with random daytime samples and also because the rate of increase in lead concentration is much smaller in the first draw region of the stagnation curve. Less replicates would therefore be required to determine mean first draw lead levels at a particular house.

Fully flushed samples† are only representative of the lowest tap water lead levels and for this reason are not normally used in population exposure surveys.

Realisation of the extremely important contribution that stagnation time makes to the overall variation in tap-water lead levels leads to the concept of *fixed stagnation time samples*. These samples are taken following a constant period of stagnation after prolonged flushing of the tap. This method effectively eliminates most of the within-house variation in lead concentration making the samples highly reproducible. If the constant stagnation period is selected to be close to the typical daytime household stagnation period these samples will also be reasonably representative of typical drinking water lead exposure levels at a particular house.

Another method of estimating exposure to lead in drinking-water is *composite proportional sampling*[7]. A consumer-operated device is fitted to the drinking water tap which splits off a small constant proportion (5%) of every

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* The consumers take these samples first thing in the morning before water has been used anywhere in the house and without flushing the tap beforehand.

† Samples taken after prolonged flushing of the tap (at least 5 service pipe volumes) at around 4 l min^{-1}.

volume of water drawn for drinking purposes*. These samples are pooled during collection so that only a single sample needs to be analysed. The lead concentration determined in this sample represents the mean drinking water concentration weighted by volume drawn for consumption. If the concentration is multiplied by the total volume of water drawn for consumption (calculated from the volume of sample collected), an estimate of total lead intake over the period of collection can be obtained. This estimate does not take into account the fact that not all the water drawn for consumption is actually consumed. Unfortunately, the proportional sampling device in its current form is fairly expensive to install and often causes considerable nuisance to the consumer.

2. INDIRECT ASSESSMENT OF EXPOSURE

We have seen that the sampling methods currently used to assess exposure levels to lead in drinking water often require large numbers of samples and only give *one measure* of exposure, for example, a mean of random daytime samples taken at a particular house. It was, therefore, decided to investigate an approach based on mathematical modelling techniques which depends on making a number of assumptions, but gives the *complete distribution* of lead concentrations in the volumes of water consumed rather than just a single summary statistic.

A brief outline of the important steps in developing the model is given below:-

(i) A simple mathematical function (stagnation curve) is used to describe the experimentally determined relationship between stagnation time of water in lead service pipes and the concentration of lead in tap-water.

(ii) Assumptions are then made about the form of the probability distributions followed by random daytime and first draw stagnation intervals.

(iii) These theoretical probability distributions for random daytime and first draw stagnation *intervals* can then be combined with the

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* Originally developed by Rijksinstituut voor Drinkwatervoorziening, Leidschendam, The Hague, Netherlands.

stagnation curve to produce separate probability distributions of daytime and first draw drinking water *lead concentrations*.

(iv) The random daytime and first draw water lead distributions are then combined by weighting each distribution by the amount of water consumed in that sample category. The first draw distribution thus receives a very low weighting, as the proportion of water consumed in this category has been shown to be small[6].

(v) Any summary statistic such as mean, median or 95 percentile lead concentrations can then be derived for the combined distributions.

2.1. STAGNATION CURVES

Experiments conducted by the WRC on three *in-situ* lead pipes in different parts of the UK have shown that the increase in lead concentration with increasing stagnation time of water in the pipe is well approximated by a log-hyperbolic function:-

$$\log_e [Pb] = \alpha - \frac{\beta}{(T+\theta)}$$

where T is the stagnation time and α, β and θ are parameters which must be estimated for each pipe.

Samples taken after prolonged flushing represent the minimum or 'base level' steady state lead concentration obtainable in water from a tap. During the first few hours of stagnation the lead concentration rises rapidly from the minimum flushing value, levelling off to an upper steady state level after about 10-20 hours.

The basic stagnation curve model has been shown to hold for large differences in water quality.

2.2. STAGNATION INTERVALS

(i) Random daytime

If the assumption is made that daytime water uses are independent and occur randomly throughout the day, then it can be shown that the *intervals* between successive uses have a negative exponential distribution. Data from a small number of different households of WRC staff, in which water use times have been recorded, support this hypothesis. The mean random daytime stagnation interval of a typical 3 person household was estimated to be about 30 minutes.

(ii) First draw

Overnight stagnation intervals were treated separately because a source of data was available from which a reasonably representative UK distribution could be derived. The occupants of each of 700 households visited during the UK Regional Heart Study (RHS)* were asked to record the time of taking an early morning first draw sample. They were also asked to record the time that water was last used in the house the previous evening. The frequency distribution of the time differences between these two observations (i.e. the overnight stagnation periods) was found to be somewhat negatively skewed, but for the purpose of this study, since so little first draw water is consumed it was considered reasonable to make the assumption of normality. The mean overnight stagnation interval was estimated as 7.3 hours, with a standard deviation of 1.4 hours.

2.3. THE EXPOSURE MODEL

Figures 1 and 2 illustrate the two stages in the development of the population exposure model.

Figure 1 shows how the continuous probability distribution functions that were fitted to the sample data obtained for the random daytime and first draw stagnation intervals were combined with a stagnation curve function to produce separate theoretical probability distributions for random daytime and first draw lead concentrations.

The random daytime and first draw distributions were then combined by weighting the probabilities with respect to estimates of the relative volumes of drinking water consumed by the population of Great Britain in each category. These estimates were obtained from a recent survey of drinking water consumption patterns in Great Britain[6] which subdivided the estimated total volume of tap-water consumed at home by the UK population into the

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* The Regional Heart Study is a long-term prospective epidemiological survey designed to investigate the geographical variations in heart disease mortality rates in the UK. The Water Research Centre is co-operating with the Royal Free Hospital's Department of Clinical Epidemiology and Social Medicine in sampling selected households for determination of trace elements in drinking water.

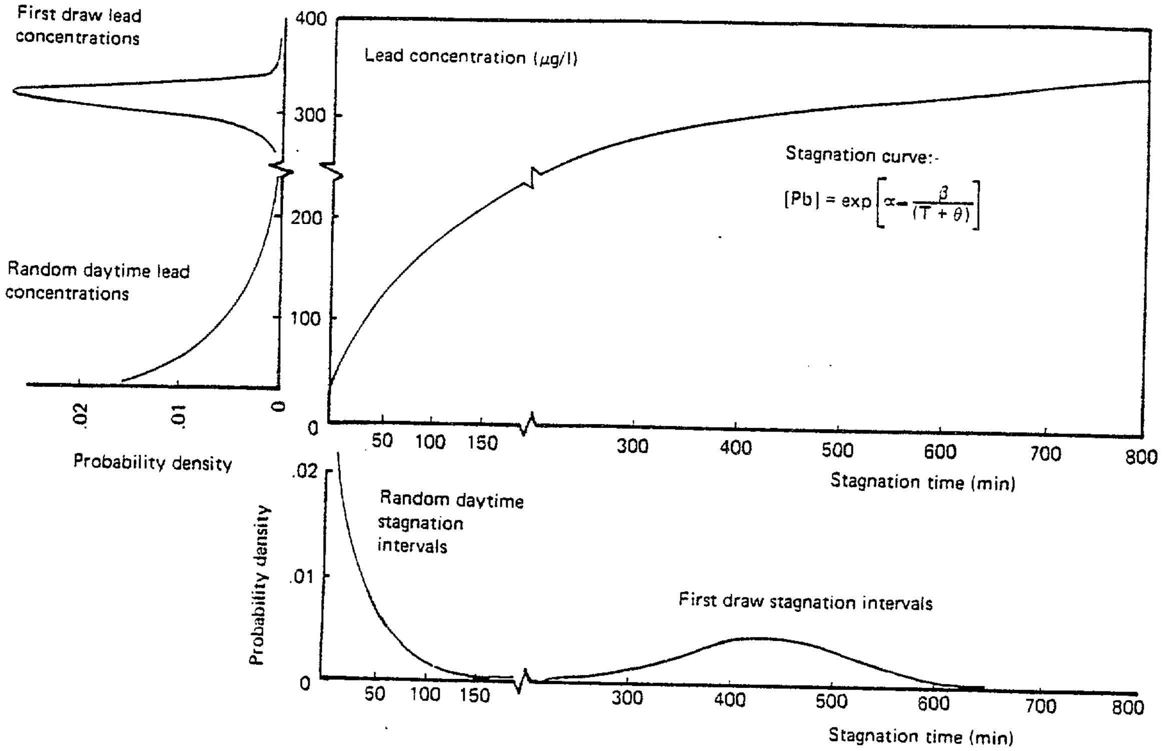

Fig. 1. Modelling the random daytime and first draw probability distributions of tap-water lead concentrations

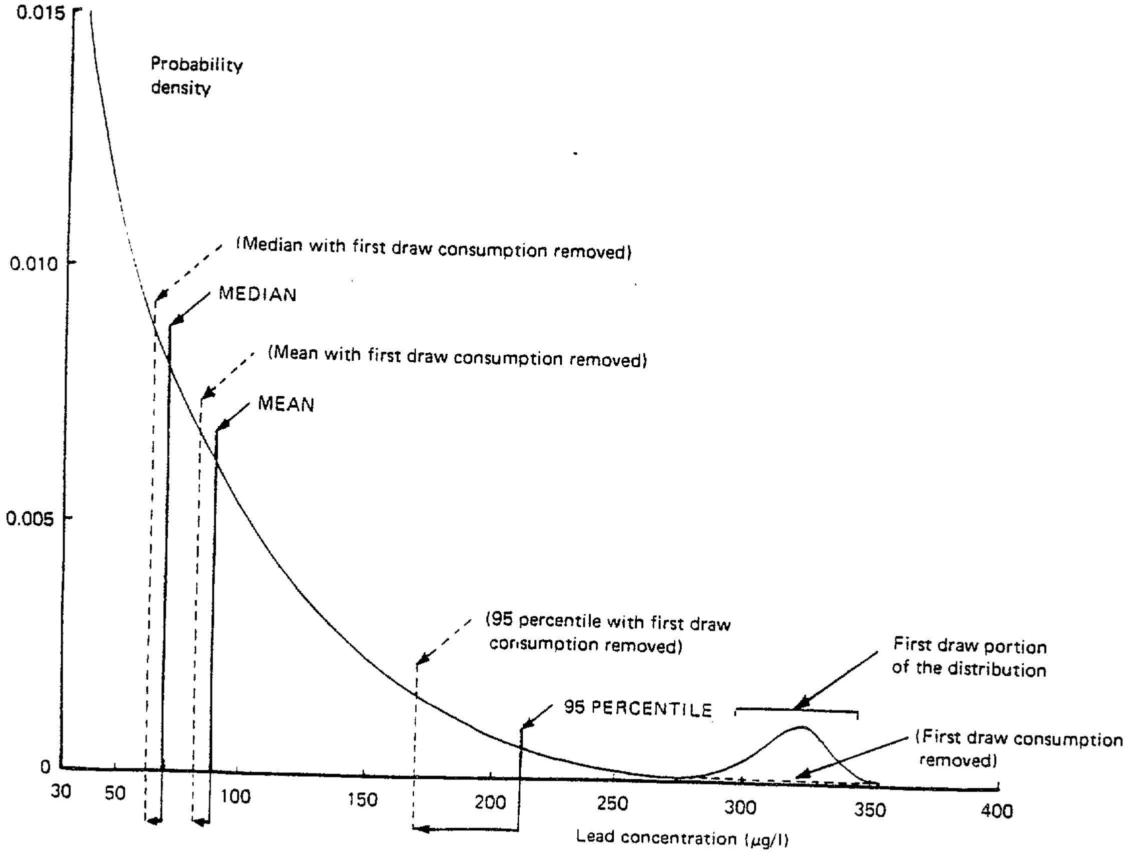

Fig. 2. Resulting theoretical distribution of drinking water lead concentrations (also showing the effect of removing first draw consumption on the mean, median and 95 percentile values (Section 2.4)

proportions of water drawn in the first draw, random daytime and fully flushed sampling categories as follows:-

First draw	4%
Random daytime	84%
Fully flushed	12%

For the purposes of this paper, the random daytime and fully flushed categories have not been separated so that the proportion of random daytime water consumed is taken as 96% of the total. Figure 2 shows the shape of the resulting theoretical distribution of drinking water lead concentrations.

It can be seen that this weighting has the effect of greatly reducing the probability of obtaining the higher first draw concentrations, because so little water is actually drunk in that category. The very small proportion of first draw water drunk results from the fact that water is normally drawn for some other purpose in the morning before being drawn for drinking. Also there is, of course, only one opportunity during the day for consuming first draw water, compared with numerous opportunities for drinking in the random daytime category.

In its present form the model gives an estimate of the distribution of lead concentrations in all volumes of water drawn from the service pipe of a *particular house* to which the stagnation curve applies. If every volume of water drawn from the service pipe has an equal probability of being consumed, then this is also the distribution of lead concentrations in all volumes of water consumed in the house. The individuals exposed to this distribution of lead concentrations are taken to be the members of a *single family unit* which was considered to be typical of all 3 person (2 adults + 1 child) family units.

As with all modelling, the model described in this paper depends on assumptions about the behaviour of the system being studied. The validity of the main assumptions made in the current model will now be briefly discussed:-

(i) Stagnation curves

The major assumption in this part of the model is that of a single stagnation curve for a particular water quality. Little is known at present about how stagnation curves vary between lead pipes fed with the same water. More work is required to assess the variability of stagnation curves between pipes in areas of constant water quality before the model can be extended.

(ii) Stagnation intervals

It is clearly not valid to assume that the negative exponential distribution of random daytime stagnation intervals shown in Fig. 1 holds for a population, as it is based on just one particular household. However, the assumption was made that the distribution of this household was typical of the distributions of households of similar sizes. More data is required from a wide range of households of different sizes in order to test the more general validity of a negative exponential distribution. The distribution of overnight (first draw) stagnation times also shown in Fig. 1 is probably reasonably representative of the UK as a whole, because the sample was obtained from a large number of households of widely differing sizes in various parts of the UK.

(iii) Pipe flushing

The model assumes that each water use completely flushes the service pipe and house plumbing so that the base level lead concentration is restored before each stagnation period. This assumption is only justified for shorter than average service pipes. In its present form the model will, therefore, tend to underestimate lead levels for longer service pipes.

(iv) Volumes of water consumed

Data is not yet available to test the validity of the assumption that the volume of water drunk is independent of stagnation time, but if for example, larger volumes of water tend to be drunk after longer stagnation periods, the model will underestimate population exposure levels.

2.4. PRACTICAL APPLICATION OF THE MODEL

The population exposure model described in the previous section can be used to assess the effect on drinking water lead levels of changing the variables in the model.

As examples of how the model might be applied in practical situations concerned with reducing exposure to lead in drinking water, consider the following questions:-

(i) What would be the effect on drinking water lead levels of persuading all consumers to flush their taps first thing in the morning before drinking?

(ii) What would be the effect on drinking water lead levels of persuading all consumers to flush their taps *every time* before drawing water for drinking.

In the first case the model shows that, although the high lead levels would be considerably reduced, the reduction in the average lead levels (mean or median) would be very slight. This is shown for the distribution of drinking water lead concentrations in Fig. 2. If first draw water consumption could be removed completely (broken line) by persuading all consumers who drink first draw water to flush their taps thoroughly beforehand, the 95 percentile point would be reduced from 210 $\mu g\ l^{-1}$ to 170 $\mu g\ l^{-1}$. The corresponding reductions in median and mean levels would, however, be much smaller: from 72 $\mu g\ l^{-1}$ to 63 $\mu g\ l^{-1}$ and from 89 $\mu g\ l^{-1}$ to 82 $\mu g\ l^{-1}$ respectively. Flushing the tap has the effect of converting the first draw concentrations into fully flushed concentrations, which then appear at the extreme left hand side of the distribution.

In the second case, *all* concentrations would be reduced to fully flushed concentrations (about 30 $\mu g\ l^{-1}$) and this would result in a large reduction in average lead levels.

2.5. POTENTIAL APPLICATION OF THE MODEL

It is clearly impossible to construct a detailed stagnation curve for every property in a water supply area. However, if more were known about the distribution of household stagnation times and also about the range of different stagnation curves which are likely to be encountered, it would be possible to design and justify a sampling procedure based on fixed stagnation time samples. The results of such a sampling procedure could be directly related to the lead intake of 'typical occupants' of each house sampled.

The use of this kind of sampling would give very much more effective information for the same amount of sampling and analytical effort required for random sampling. A fixed stagnation time sampling scheme could be used to assess the behaviour of the service pipe and plumbing at an individual house or used in a sample survey over a water supply district. If used on a district basis, the fixed stagnation time sampling would reveal the statistical distribution of individual mean intakes thus providing a much firmer basis for deciding what action may be necessary to reduce lead levels than can be achieved with random sampling.

However, further investigations are required to extend the currently available information on stagnation curves and stagnation intervals before an approach based on fixed stagnation time sampling can be recommended.

3. CONCLUSIONS

1. The problem of the variability of tap-water lead levels can be approached by separating its two main components:-

    (i) The behaviour of the service pipe and household plumbing (stagnation curve).

    (ii) The behaviour of the consumer (water use patterns).

This paper has demonstrated the feasibility of predicting distributions of lead concentrations by combining stagnation curve information with information on household water use patterns.

2. In its present form, the model is capable of predicting the effect on drinking water lead levels of changes in water use behaviour.

3. The modelling approach outlined in this paper offers potential justification for fixed stagnation time sampling methods which would be far more informative than the currently used random sampling procedures.

ACKNOWLEDGEMENTS

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REFERENCES

1. R.G. Ainsworth, R.J. Bailey, B.T. Commins, R.F. Packham, and A.L. Wilson. *Lead in Drinking Water*. WRC Technical Report TR 43, 1977.
2. G.F. Craun, and L.J. McCabe, *Problems Associated with Metals in Drinking Water*. Journal AWWA, November 1975, 593-599.
3. J.M. Carter. *Lead in Drinking Water*. Water, September 1977, 2-5.
4. Department of The Environment. *Lead in Drinking Water: a Survey in Great Britain 1975-1976*. HMSO Pollution Paper No. 12, 1977.
5. S.J. Pocock. *Factors influencing Household Water Lead: a National Survey*. Archives of Environmental Health, 1980, *35* (1), 45-51.
6. S.M. Hopkin, and J.C. Ellis. *Drinking Water Consumption in Great Britain*. WRC Technical Report TR 137, 1980.
7. B.J.A. Haring. *Human Exposure to Metals released from Water Distribution Systems, with particular reference to Water Consumption Patterns*. Tribune CEBEDEAU, 1978, *419*, 349.