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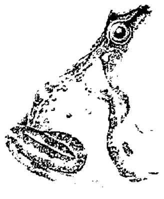

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PARTICULATE LEAD IN WATER SUPPLIES (TMU 9024)

Final Report to the Department of the
Environment - May 1987 to December 1991

DoE 2963 - /1

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Report No: DoE 2963

April 1992

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Contract Manager: S M Williams

Contract No: TMU 9024

Contract Duration: May 1987 - December 1991

DoE Reference No: PECD 7/7/230

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DoE Nominated Officer - 25 copies

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PREFACE

Elevated lead levels in drinking water can be a problem in certain areas of the UK. Lead occurs as particulate and soluble forms. Remedies for soluble lead have been developed and applied. Areas experiencing particulate lead did not respond to these remedies and further investigations were required.

The Department of the Environment placed a contract (PECD 7/7/230) with WRc in May 1987 to conduct a study on the occurrence of particulate lead and the development of suitable remedial action.

This final report presents the work undertaken.

SUMMARY

There is a legal requirement that the UK Water Industry will achieve compliance with the Water Supply Regulations (1989)[1]. For lead this is a requirement to achieve a concentration of no more than 50µg/l in all samples analysed for lead. In the case of soluble lead, problem areas have been identified, and successful remedies have been developed. However, insoluble particles of lead corrosion products have been identified in tap water at various locations throughout the UK. These occur despite the application of remedies developed for soluble lead problems.

The objectives of the work described in this report were to confirm the occurrence of particulate lead and to determine the circumstances of its occurrence. Remedial measures were investigated.

Particulate lead problems were evident throughout the UK, generally associated with lead-plumbed premises supplied from soft upland sources.

A size fractionation technique was developed and two types of particulate lead were identified using this technique; flaking lead and particulate/colloidal lead.

Field trials have shown that elevated particulate lead levels are associated with elevated levels of iron and organic matter. Reducing the levels of these parameters results in a decrease in the incidence of particulate lead. The results of the limited field trials to assess the effect on lead of CO_2 cleaning lead service pipes show that this technique can eliminate particulate lead under certain circumstances.

Steps should be taken to reduce levels of iron and organic matter in water supplies that experience particulate lead problems in distribution. More comprehensive long-term trials to investigate the effectiveness of CO_2 cleaning lead service pipes are recommended.

1. INTRODUCTION

One of the requirements on the UK Water Industry is to comply with standards of drinking water quality set out in the Water Regulations. These state that, in the case of lead, concentrations in tap water samples should not exceed 50µg/l. Where this target is not met, two different types of problem have been identified. Commonly, high soluble lead levels are found and these have been largely overcome by water treatment to reduce plumbosolvency. However, in some areas, such water treatment is unsuccessful. In these areas, the problems may be due to particulate or flaking lead. This project was therefore initiated to investigate the problem of particulate lead. Specifically, the aims of the project described in this report are:

(1) To confirm the occurrence of particulate (insoluble) lead in areas which do not comply with the requirements for lead set out in the Water Supply Regulations.

(2) To determine circumstances under which particulate lead occurs and seek critical factors causing this form of lead contamination.

(3) To investigate and validate potential remedial action.

This report summarises the findings from a national survey performed to assess the extent of particulate lead problems. A description is given of the development of an analytical method for differentiating between soluble and particulate lead. A number of field and laboratory trials performed to identify the factors that affect particulate lead are described. Finally several investigations of remedial action to reduce particulate lead problems are described.

2. OCCURRENCE OF PARTICULATE LEAD: NATIONAL SURVEY

During the period 1983 to 1985 the Water Authorities in England and the Welsh and Scottish Offices carried out surveys specified by the Expert Advisory Group on Identification and Monitoring (EAGIM surveys)(1) in their supply zones, to identify priority areas for remedial action for reducing lead levels. Priority areas were defined as those where more than 2½% of random daytime samples exceeded lead values of 100µg/l, the EC guideline for lead then applying. In the EAGIM survey total lead concentrations were measured, no distinction was made between soluble and particulate lead.

The current contract undertook to collect further data from water authorities to:

(i) investigate the extent of the particulate lead problems;
(ii) identify causes of and circumstances promoting the occurrence of particulate lead;
(iii) find suitable sites for further investigations involving fieldwork.

As many data as possible were collected covering EAGIM surveys, effects of remedial actions, complaint data and special case studies.

All water authorities in England and Wales and some of the Scottish regional councils were approached for information on performance in the EAGIM surveys and the occurrence of particulate lead.

The national survey identified areas with a substantial incidence of lead concentrations above the EC guideline of 100µg/l lead maximum then applying. As no distinction was made between soluble and insoluble lead problems during the EAGIM surveys, it has been assumed that where treatment to reduce plumbosolvency has failed to reduce lead levels, particulate lead is implicated.

An analysis of all available data indicated that particulate lead problems are most likely to occur in areas that are supplied by soft upland water with a low alkalinity, low in hardness, often high in colour and organics. Often these supplies receive only minimal treatment, resulting in high iron concentrations and occasionally high manganese and aluminium concentrations and poor pH control. Table 2.1 summarises a typical composition of a water with potential particulate lead problems.

Table 2.1 - Typical composition of a water with potential particulate lead problems

Alkalinity	<50mg/l as $CaCO_3$
Hardness	<50mg/l as $CaCO_3$
Total organic carbon	>2mg/l as C
Colour	>10 - 20 Hazen
pH	7.8 - 9.0
Iron	often high
Manganese	
Aluminium	

This implies that approximately a third of the water supplies in the United Kingdom has potential particulate lead problems where lead pipes or lead-lined storage tanks are present.

Although hard water areas, high in alkalinity, are more likely to suffer from soluble lead problems, particulate lead problems might also occur where the corrosion layer inside pipes is disturbed either mechanically or by chemical changes in water quality.

3. SIZE FRACTIONATION ANALYSIS

Lead can exist in natural waters in a great variety of physicochemical forms, including free aquo ions, inorganic ion-pairs and complexes, organic complexes, associations with highly dispersed colloidal material, as suspended particles of insoluble salts, or adsorbed on inorganic particulates. These various species can be classified on the basis of size distribution, as shown in Table 3.1, and size fractionation procedures can therefore prove extremely useful in investigations of lead speciation.

Table 3.1 - Classification scheme for lead species in water (adapted from Fielding and Hunt[(2)])

APPROX. SIZE (nm dia, log scale)	10000----------	----------	100--------	10----------	1----------	----------
CLASS	Minerals and precipitates	Adsorbed on inorganic particulates	Highly dispersed colloidal material	Bound to macro-molecules	Organic chelates, other inorganic ions, ion pairs and complexes	Free aquo ions
EXAMPLES	$PbCO_3(s)$	Adsorbed on hydrous iron and manganese oxides and clay minerals	Adsorbed on hydrous iron and manganese oxide colloids	Pb fulvic acid complexes	Pb-EDTA $PbCO_3$	Pb^{2+}

Harrison and coworkers(3) have described a scheme involving filtration to five different size fractions coupled with measurements of "labile" lead as defined by anodic stripping voltammetry and Chelex chelating ion-exchange resin techniques. This basic approach was considered appropriate for the present study and a similar filtration scheme was adopted on the initial sampling programme. This involved filtration of the tapwaters through Nuclepore polycarbonate filters of pore sizes 12, 1, 0.4, 0.08 and 0.015µm and determination of lead, iron, manganese, aluminium and total organic carbon (TOC) in each fraction. Polycarbonate filters were preferred to cellulose acetate or nitrate membranes because they provide better size discrimination at small pore sizes (Laxen and Chandler(4)) and are less prone to adsorption problems (Gardner(5)).

Various trials were performed with the different pore size filters in order to establish, and be able to correct for, the percentage adsorption of lead on the filters.

The method development and description are given in Appendix A. The outcome of the development work was a method for identifying the proportion of lead, iron, manganese, aluminium and total organic carbon in five size fractions of particles ranging from >12 to <0.015µm in diameter. Using this method as part of the project it has been possible to identify whether a particulate lead problem exists in a given area, and whether the particulate lead is chemically associated with other constituents of the water (such as iron, aluminium etc.) in the same size fraction.

4. FIELDWORK: CONFIRMATION OF PARTICULATE LEAD PROBLEMS AND INITIAL INVESTIGATION OF CAUSES

4.1 Definition of particulate lead

Two types of particulate lead problems have been identified in the areas studied, "flaking" lead and "particulate/colloidal" lead.

Flaking lead problems are caused by large black particles visible to the consumer. These resemble "tea leaves" or "black specks" in the water. Size fractionation analysis (see Section 3) has shown that flaking lead mainly consists of particles >12µm. Flaking lead problems have been identified in Areas A, B, D and K. Although this has not been confirmed by size fractionation and sampling as part of this project, the occurrence of flaking lead has also been noted in other parts of the UK. Particulate/colloidal lead problems are caused by smaller particles than those responsible for flaking lead problems, and particles are generally not visible to the consumer. Size fractionation analysis has shown that most particles are present in the size fractions between 0.08 and 12µm. Particulate/colloidal lead problems are mostly associated with "dirt" in the water and a correlation has been found between lead and iron in the water. Identified particulate/colloidal lead zones are Areas C and L.

There is no clear distinction between the two types of lead problem as large particles can occasionally be found in particulate/colloidal lead areas possibly as a result of disturbance of the corrosion layer.

4.2 Occurrence of particulate lead in Area A

Area A has suffered an intermittent lead problem in the past. This area is supplied by three surface sources, all of which are soft, have a low alkalinity and relatively high pH. The supplies are mixed depending on demand and availability. Each time a change in source occurs, flow reversals can take place in feeder mains and in some smaller distribution mains with a possibility of increased particulate matter in the conveyed water. The water is not plumbosolvent.

A large sample of 190 premises, comprising 75% known lead-plumbed houses and 25% other or unknown type of pipe material, was monitored. These premises were located inside and outside the problem area and were all supplied with water of the same type.

The sampling programme generated data showing a significant correlation between lead and iron and aluminium. Lead problem areas showed greater iron and aluminium concentrations compared with non-problem areas, although premises with non-lead pipes within the problem area did not register increased iron and aluminium levels.

A detailed investigation of selected properties demonstrated that hydraulic circumstances have no significant effect on metals concentrations in tap water from premises outside the problem area. In problem properties, flushing and water hammer tended to increase the levels of lead, iron, manganese and aluminium. Stagnation in lead pipes did not significantly alter metal concentrations. A high proportion of the metals in these samples was present in the particulate form, except aluminium which was mainly present in solution.

It is believed that the appearance of lead flakes and particulate matter is due to the destabilisation of the lead corrosion layer, which contains iron, aluminium and manganese, as a result of some chemical or mechanical disturbance. Examples of the former type are changes in water quality and the effects of temperature, which can cause changes in the rate of chemical reactions in addition to the physical effects of pipe bending and cracking of corrosion layers due to expansion and contraction. The destabilised layer flakes off when water velocity is high or when water hammer occurs. The absence of any effect of 30 minutes stagnation on metals levels supports this. The occurrence of lead problems was not associated with a deterioration in water quality, since no differences in water quality inside and outside the problem area were detected. It is however possible that differences existed in the recent past. No relationship was found between elevated lead levels and TOC or any other water quality parameter (over the ranges operating) excepting the positive correlations between lead and the metals iron, aluminium and sometimes manganese.

Subsequent studies have indicated that the lead problem in Area A is intermittent. High lead levels were not always reproducible in the problem area.

A detailed discussion on the investigation of particulate lead in Area A is presented in Appendix B.

4.3 Occurrence of particulate lead in Area B

The type of particulate lead problem in Area B is one of flaking lead where the particulate material is visible. The flakes are thought to be parts of an exfoliating lead corrosion layer. Area B experiences a seasonal pattern of flaking lead, flakes being much more numerous in the summer months. An analysis of samples taken in response to consumer complaints indicated a relationship between flaking lead at concentrations of >100µg/l and other metals, especially iron and aluminium, but also to a lesser extent copper and manganese.

Random spot sampling was conducted in Area B to investigate further the occurrence of flaking lead. The results of this exercise confirmed that the lead problem in Area B is one of flaking lead with particles >12µm in size which are visible to the consumer. In addition, lead problems are generally associated with high flows and high demands. Despite the two sources and associated blends in supply in this area, lead problems occurred throughout the study area. Long-term monitoring was carried out at 8 properties in Area B at fortnightly intervals for six months between March and September (i.e. over the summer period). Samples were taken at high and low flows and analysed for a range of chemical parameters including alkalinity, turbidity, TOC, hardness, pH, conductivity, free and total chlorine, iron and aluminium. Note was also made of the number of flakes present in the samples.

The purpose of the work was to investigate the factors that affected the occurrence of particulate lead. In general, many more flakes were present in the fast flowing sample, indicating that there is a definite hydraulic effect. The only water quality parameter that showed an association with lead was iron. In general elevated lead concentrations were associated with elevated iron concentrations.

4.4 Analysis of lead corrosion products

During the initial stages of the project several areas which suffer from problems of particulate lead had been identified. In general these areas received soft upland supplies which also sometimes contained organic matter. Iron was generally present in the supply either from source or from mains corrosion. However there are areas receiving water with apparently similar characteristics that do not experience particulate lead problems. To investigate this apparent anomaly, lead pipes were exhumed, representing many different water supply areas, some experiencing and some not experiencing particulate lead problems as a result of lead service pipes. A summary of samples investigated is presented in Table 4.1.

Table 4.1 - Location of pipe samples for corrosion product analysis

AREA	NUMBER OF SAMPLES EXAMINED	NATURE OF LEAD PROBLEM
A	8	Flaking lead
B	2	Flaking lead
E	2	Particulate/colloidal lead
M	3	Historical flaking lead
N	1	Non-problem area
O	1	Particulate/colloidal lead
P	6	High aluminium
Q	7	Historical flaking lead
R	1	Non-problem area
S	1	Flaking lead
T	1	Non-problem area
U	1	Particulate/colloidal lead

The corrosion products were scraped off the pipes by hand, after first brushing away any loose material from the surface. The corrosion products came away in a powder form and were filtered through a 0.250µm filter. The coarse material comprised exclusively lead and was thought to be uncorroded lead pipe disturbed during the scraping process. For this reason, the material >0.25µm was excluded from the analysis. Both visual and chemical analyses were made.

Visual inspection of the pipe surfaces and associated lead corrosion products showed that most samples consisted of two layers. The bottom layer was fairly similar in all pipes, being creamy white in colour and well attached to the surface of the pipe. Chemical analysis showed this layer to be very high in lead and inorganic carbon, probably a lead carbonate deposit formed on the metal surface. This layer had very low levels of other metals and is possibly the first product of the reaction between lead pipes and water containing the carbonate anion.

A summary of the chemical data obtained from the upper layer of the corrosion product is shown in Table 4.2. The composition was found to vary with location and it appears to be the nature of this upper layer that determines the nature of the lead problem. The upper layer had a high lead content but varied in the levels of other metals and in organic carbon. On a weight basis, lead was generally responsible for over 50% of the corrosion product.

Examination of the chemical data also shows that there is a high variability in the composition of corrosion products taken from the same supply area. Localised variations in water quality may have contributed to this, as well as variabilities introduced during sampling and analysis.

Some general trends could be identified. Where particulate/colloidal or flaking lead is experienced, the upper corrosion layer contains high levels of organic carbon, iron and aluminium and occasionally manganese. In non-problem areas, corrosion products consist of lead and organic carbon to the exclusion of most other metals.

The samples from Area P were exceptions containing on occasion only a small proportion of lead and high levels of aluminium and organic carbon, the corrosion layer being very thick and smooth. These results are very different from those measured elsewhere and it is thought that this is due to the presence of aluminium salts as a result of the high aluminium levels in the source water.

It would appear that problems with lead in drinking water can vary depending on the nature of the upper layer of corrosion product. This is probably controlled by water quality and mains material. It may therefore be possible by considering these factors to identify areas where flaking or particulate/colloidal lead may be expected to be a problem.

The most noticeable correlation in the corrosion products examined is the relationship between lead, iron and manganese and organic carbon. It seems likely that lead dissolved from the service pipes may become associated with organic or metalliferous deposits, either by adsorption onto manganese or iron accumulations or by complexation with organics. The results suggest that it may be the combination of metals with organic carbon that exacerbates particulate lead problems since corrosion products from non-problem areas still contain organic carbon but are not necessarily associated with other metals.

A visual comparison has been made between the corrosion layers of lead pipes suffering flaking lead and those on pipes suffering particulate/colloidal lead problems. These were found to differ. Particulate/colloidal lead problems were associated with smooth deposits, grey/orange/brown in colour. Flaking lead arose from thick deposits, black/brown in colour and flaky in appearance.

Table 4.2 - Results of analysis of lead corrosion products. Iron, manganese, aluminium and organic carbon levels (in gmoles) as a % of lead levels (in gmoles)

AREA	SAMPLE	% LEAD	% IRON	% MANGANESE	% ALUMINIUM	% TOC
A	1	100	25	0	12	93
	2	100	2	0	2	43
	3	100	20	1	17	51
	4	100	4	0.3	6	51
	5	100	55	1	22	27
	6	100	5	0.3	5	124
	7	100	6	0.3	6	41
	8	100	-	-	-	39
B	1	100	193	157	92	432
	2	100	69	29	37	263
E	1	100	140	0.8	22	324
	2	100	12	0.4	7	150
M	1	100	19	0.3	2	246
	2	100	1	0	0.6	71
	3	100	0.5	0	0.3	50
N		100	9	0	0.3	12
O		100	56	5	24	63
P	1	100	150	20	200	950
	2	100	2	0.9	9	84
	3	100	11	9	38	129
	4	100	6	2	36	-
	5	100	67	5	130	711
	6	100	6	0.6	14	86
Q	1	100	2	0	0.3	66
	2	100	2	0	0.3	68
	3	100	22	0.3	5	145
	4	100	40	0.8	13	147
	5	100	1	0	0.8	55
	6	100	42	0.4	13	347
	7	100	2	0	1	80
R		100	17	2	3	27
S		100	72	77	100	340
T		100	3	2	10	74
U		100	35	2	25	100

4.5 Summary of initial findings

Two types of particulate lead have been identified using size fractionation studies; flaking and particulate/colloidal lead. Detailed studies have identified a number of areas in the UK where problems of this type occur.

Particulate lead is often associated with internal lead corrosion layers containing high levels of organic carbon and other metals, e.g. iron, aluminium and occasionally manganese. Non-problem areas provide service pipes with internal lead corrosion layers containing organic carbon but negligible levels of metals. These compositional factors may affect the stability of internal corrosion layers and thus may account for the difference in behaviour. Corrosion deposits in flaking lead pipes are differentiated physically from particulate/colloidal lead pipes by the much rougher, flaking surface.

5. FIELDWORK TO IDENTIFY CAUSES AND SOLUTIONS

5.1 Area C pipe rig

5.1.1 Introduction

Water supplied to Area C originates from a soft upland source rich in iron, organic matter and sometimes manganese.

For a number of years this area had failed to meet the requirements of the EAGIM lead surveys and attempts were made to treat the water (pH correction and orthophosphate dosing) to control lead levels. These attempts were unsuccessful. A lead pipe rig was therefore constructed and set up at the water treatment works supplying Area C. The experimental programme had the following objectives:

- To identify the circumstances that affect lead concentrations in Area C tap water;

- To test the effects of remedial actions.

Water from the treatment works fails to comply with EC limits for iron and occasionally manganese; a new treatment plant is therefore planned. A pilot plant was commissioned to treat a small proportion of the water to remove iron, manganese and organic matter and to increase the alkalinity slightly from <10mg/l to ~30mg/l as $CaCO_3$.

The pipe rig was initially used with "old" treated water and was subsequently moved to the pilot plant to compare the effects of the pilot plant water and the "old" water on lead levels.

5.1.2 Experimental plan

Table 5.1 summarises the experiments performed during the pipe rig trials. Each experiment entailed taking a flushed sample from all 8 pipes on the rig, then taking samples after ½ hour, 2 hours and 18 hours stagnation so a lead concentration versus time stagnation plot could be obtained. When the pipe rig was moved to the pilot plant, physical disturbance of the lead pipes led to erratic results; limited sampling was carried out at fortnightly intervals to assess when the rig had stabilised. Stagnation experiments on the new and old water subsequently proceeded.

5.1.3 Results and conclusions

A full description and analysis of the pipe rig results are contained in Appendix C.

The overall conclusions reached are that the factors that affect particulate lead concentrations in Area C tap water are the joint presence of particulate/colloidal iron and organic matter, and physical disturbance of the internal corrosion layer. Treating the source water to remove the iron and organic matter results in a significant reduction in lead levels. This reduction in lead concentrations is unaffected by variation in alkalinity or pH or by the addition of silicates and phosphates.

Table 5.1 - Timetable of experiments with the lead pipe rig at the treatment works supplying Area C

DATE		CHEMICALS ADDED	ALKALINITY (mg/l $CaCO_3$)	pH	COMMENTS
OLD WATER - HIGH IRON AND ORGANICS, LOW ALKALINITY					
Aug 88		-	<10	8.4	Intermittent water flow
Aug 88 and April 89		-	<10	8.3 - 9.5	Repeated 4 times to get baseline
Aug 88		$FeCl_3$ + NaOH to give $Fe(OH)_3$ (1mg/l as Fe)	<10	7.2	Colloidal iron
April 89		$FeCl_3$ (0.5mg/l as Fe)	<10	7.9	Soluble iron
April 89		$FeCl_3$ (0.8mg/l as Fe)	<10	7.2	Soluble iron
April 89		$FeCl_3$ (0.5mg/l as Fe)	<10	9.6	Soluble iron
May 89		polyphosphate (2mg/l as P_2O_5)	<10	9.4	Kalipol
NEW WATER (LOW IN IRON AND ORGANICS) IN FOUR PIPES ('NEW' PIPES). OLD WATER IN FOUR PIPES ('OLD' PIPES)					
Sept 89	New	-	30	9.0 - 9.9	Duplicated to get baseline
Sept 89	New	Reduced CO_2 and lime	21	8.6 - 9.6	
Sept 89	New	$Fe(OH)_3$ (0.25mg/l as Fe)	30	8.7 - 9.5	Colloidal iron
Sept 89	Old	$Fe(OH)_3$ (500mg/l* as Fe)	<10	7.9 - 8.5	Colloidal iron
Oct 89	Old	$Fe(OH)_3$ (1.28mg/l as Fe)	<10		Colloidal iron
	New Old	orthophosphate (0.8mg/l as P)		8.2 - 8.7 9.4	

*an unintentional overdose of $Fe(OH)_3$ entered the four pipes supplied with old water.

Table 5.1 continued

NEW WATER (LOW IN IRON AND ORGANICS) IN FOUR PIPES ('NEW' PIPES).
OLD WATER IN FOUR PIPES ('OLD' PIPES)

DATE		CHEMICALS ADDED	ALKALINITY (mg/l $CaCO_3$)	pH	COMMENTS
Oct 89	New	Old water	<10	9.4 - 9.9	Testing whether pipes have a "memory" Duplicated
	Old	Old water	<10	9.4 - 9.9	
Oct-Nov 89	New	Old water	<10	8.5 - 9.0	Duplicated
	Old	Old water + CO_2 + lime	30		
Nov 89	New	New water	20	9.0	Duplicated
Nov 89	Old	Old water	<10	8.5 - 9.0	Duplicated
Nov 89	New	New water + CO_2 + lime	30	9.0	Duplicated
	New	New water + CO_2 + lime	50	9.0	Duplicated
Dec 89	New	New water + silicate (1mg/l as SiO_2)	20	9.0	Duplicated
Jan 90	New	New water + poly phosphate (2mg/l as P_2O_5)	20	9.0	Duplicated
Jan 90	New	New water + silicate (13mg/l as SiO_2)	20	9.0	Duplicated
Mar 90	New	pH varied between 7.5 and 8.5	30	7.5, 8.0, 8.5	Duplicated

5.2 Remedial actions to combat the occurrence of particulate lead in Areas D, E and F

Several areas within the north of England have been identified as experiencing problems of particulate lead. Trials have been performed in Areas D, E and F with orthophosphate and polyphosphate to try and remedy the problem. A summary of the trials is included in Table 5.2.

Table 5.2 - Summary of trials of chemical treatments intended to reduce lead levels in zones in the north of England

AREA	TYPE OF LEAD PROBLEM*	FIRST TREATMENT	DATE	SECOND TREATMENT	DATE
D	Soluble/ flakes	Poly-phosphate (Kalipol)	July 1988	Ortho-phosphate	January 1989
E	Particulate/ colloidal	Ortho-phosphate	April 1989	-	-
F	Particulate/ colloidal	Poly-phosphate (Kalipol)	May 1988	Ortho-phosphate	April 1989

* Type of problem established by size fractionation

The full results have been reported in Appendix D. The main conclusions are as follows:

(1) Areas E and F have both been identified by particle size fractionation as zones having a particulate/colloidal lead problem. Orthophosphate dosing in Area E reduced iron levels slightly; polyphosphate dosing in Area F reduced iron levels significantly. The results for lead were inconclusive; however the expected seasonal rise in lead levels did not occur during dosing in both zones.

(2) In Area D, a mixture of particulate and soluble lead has been identified. Polyphosphate was ineffective at reducing total lead levels, but orthophosphate dosing caused a significant decrease both in average total lead and in the peaks in lead concentrations. It is thought that the orthophosphate may be effective on the soluble portion of the lead in the tapwater. Iron was not monitored before dosing so effects cannot be assessed.

Throughout these trials, it was not possible to directly compare lead levels in areas with and without chemical dosing, as no control areas were monitored due to practical difficulties. Assumptions were therefore made regarding anticipated seasonal trends in lead levels.

Overall, the results indicated an association between lead and iron. Phosphates can act to reduce iron levels and this in turn caused a reduction in lead levels in Areas E and F. In Area D lead levels reduced following orthophosphate dosing, probably due to a reduction in soluble lead levels.

5.3 Remedial actions to combat the occurrence of flaking lead in Areas A and B

5.3.1 Introduction

Previous work(6) (see also Appendix C) had shown that there may be an association of particulate lead with colloidal or particulate iron. Two potential sources for this particulate iron have been suggested.

(1) Iron deposits from distribution mains may be passing into lead service pipes. Lead may adsorb onto these particles or lead flakes may be physically disturbed by the scouring action of the deposits.

(2) Soluble or particulate iron may be incorporated into a corrosion layer on the internal surface of the lead service pipe. Periodically this layer may become unstable and particles may be released into the water in the service pipe.

Further work has now been conducted in order to identify which process is responsible. This involved the fitting of filters at selected properties and extensive monitoring to assess the effects of excluding particulate matter already in the water supply, from the lead service pipe.

5.3.2 Design of surveys

In both Area A and B, 6 properties were identified which were supplied via lead service pipes and which experienced particulate lead problems. In each trial area, 3 properties were to be fitted with suitable filters, the water supplied to 3 adjacent properties would remain unfiltered to act as controls. Details of the properties chosen and the fitting and testing of the filters are reported in Appendix E.

The sampling programme involved 30 weeks of sampling beginning in summer, with water samples being obtained at low and at high flow rates. Filter cartridges were changed fortnightly to prevent clogging of the filter pores.

Sufficient results were obtained from Area B using 1µm filters to enable a statistical analysis to be performed (see Section 5.3.3 and Appendix F). Sampling in Area A was problematic. Filter blocking and consumer difficulties were encountered which seriously delayed the fitting of suitable filters and the start of sampling. Only one pair of properties (filtered and control) provided long-term data with initial results obtained in late winter. 10µm filters were used in Area A.

5.3.3 Results and conclusions of filter trials

The results of these trials are presented in detail in Appendix F, Sections 1 and 2. The conclusions can be summerised as follows.

(a) Area A

Over the 31-week sampling period, lead and iron levels were in general very low at both the control and experimental properties. In view of this virtual absence of particulate lead in Area A during the sampling period, no conclusions could be drawn on the effectiveness of filtering in eliminating particulate lead in this area.

(b) Area B

The filters do not lower concentrations of total lead in the water, although they do have an effect on the presence of particulate iron. There is a link between iron and lead. High flow samples are higher in lead and occasionally higher in iron than low flow samples. All this implies that the amount of particulate iron present in the service pipe of filtered properties is sufficient to result in the same quantity of particulate lead as in non-filtered properties. So, although filtering reduces levels of particulate iron, it is likely that particulate iron already exists on the internal surface of the service pipe and acts as a reservoir. Filtering will have an effect only on newly incoming particulate iron and not on matter already deposited in the service pipes.

5.3.4 CO_2 cleaning of service pipes

In an attempt to remove the reservoir of particulate material discussed in section 5.3.3b, CO_2 scouring was conducted on the service pipes to two properties in Area B undergoing filtration. A full statistical analysis of the results obtained for each pair of properties (control and filtered) was conducted. The results are presented in detail in Appendix F Section 3. The small scale of the survey in terms of number of premises sampled will not enable firm conclusions to be drawn but permits some comparisons to be made.

A summary of the findings of the statistical analysis is presented in Table 5.3.

Table 5.3 Effect of CO_2 scouring on particulate lead and iron levels at two properties in Area B

PROPERTY IDENTIFICATION	SAMPLE TYPE	EFFECT OF CO_2 SCOURING ON	
		PARTICULATE LEAD	PARTICULATE IRON
2	Low Flow	↓↓	→
	High Flow	→	→
4	Low Flow	→	→
	High Flow	↓↓	↓↓

Key → no significant effect
↓↓ significant decrease (95% confidence level)

In view of the generally low levels of particulate lead in low flow samples, the statistically significant decrease in this parameter measured for property No 2 as a result of CO_2 scouring is considered to be unimportant, being associated with no improvement in the already acceptable lead PCV compliance rate. The statistically significant decrease in particulate lead and iron levels at high flow rates in property No 4 as a result of CO_2 scouring is however important. A substantial improvement in the lead PCV compliance rate was achieved with the elimination of the particulate lead problem.

Therefore, these results for both pairs of properties do not provide consistent conclusions on the effect of CO_2 scouring. The property pair No 4 provided promising results with particulate lead eliminated by the scouring technique. The short post-scouring period does not permit the long-term effect of scouring to be established. The contrasting behaviour of property pair No 2 may be a reflection of possible differences in procedure of the CO_2 scouring performed at each property. The existance of such differences can however not be established.

5.4 Investigations on exhumed service pipes

5.4.1 Sampling procedure

Further investigations into the nature of the corrosion deposits within lead service pipes were undertaken in an attempt to obtain additional evidence to confirm earlier findings. The inorganic composition of deposits removed from service pipes collected from various sites in the UK was examined. Microbiological factors may also influence the behaviour of the lead corrosion layer and these were investigated using standard plate count and biofilm techniques (see below). Lastly pipe metal composition was examined. The results of these chemical and microbiological analyses were compared with the lead and iron levels of water samples collected immediately prior to the disconnection of the supply before pipe removal and with the historical behaviour in order to identify correlations between particulate lead at the tap and corrosion deposit or pipe characteristics.

Two sites within each of five distribution areas were selected representing flaking lead, soluble lead and non-problem areas. Site details are summarised in Table 5.4 and relevant source data in Table 5.5. Immediately before supply disconnection, water samples were collected as indicated below.

(i) Random daytime (total metals)

(ii) Low flow (5 l/min) (total plus dissolved metals)

(iii) High flow (full bore) (total plus dissolved metals).

The pipe was removed after disconnection and prepared for microbiological and chemical analysis.

Details of the analytical techniques are given in Appendix G, Section 1.

Table 5.4 Summary of areas selected for service pipe exhumations

Area	Source	Historical behaviour
B	Soft upland source, treated using aluminium sulphate coagulant, sedimentation, rapid gravity filtration, pH correction and chlorine	Flaking lead
C	Soft upland source rich in iron, organic matter and sometimes manganese	Soluble plus particulate lead
G	Lowland reservoir and lowland river, moderate hardness, occasionally high in iron, high total organic carbon	Elevated lead
H	River derived, moderately soft water, iron levels occasionally high	Soluble lead
I	River derived, hard water plus spring sources	No problem
J	Hard spring water with high turbidity during periods of heavy rain	No problem

Table 5.5 - Summary of source data* for areas supplying lead pipes

AREA	ALKALINITY (mg $CaCO_3$/l)	pH	TOC (mg C/l)	Fe (µg/l)
B	12	8.8	-	33
C	11	7.5	1.7	146
G§	123 121	8.6 8.5	5.6 4.8	150 55
H	82	8.1	1.7	78
I	84 - 227	7.2 - 8.6	-	10 - 410
J	199 - 260	7.2 - 7.9	-	10 - 140

- No information available.
* Mean values except Areas I and J where range is given.
§ Area G has two sources.

5.4.2 Results and conclusions

The detailed results of these investigations are presented in Appendix G Section 2. The findings can be summarised as follows

(a) Water quality at exhumation sites

One property in Area B demonstrated clearly the typical flaking lead problem well documented in this area. Both Areas C and G gave no evidence of the soluble and elevated lead problems respectively encountered in the past. This serves to show the variable nature of lead problems and their non-appearance at certain properties or at certain times within problem zones.

Particulate lead was identified in Area H, where a soluble lead problem had been anticipated and also in Area I where historically no lead problem had been encountered. The association of particulate iron with the particulate lead confirms previous findings.

(b) Pipe metal composition

All lead service pipes were almost or entirely 100% lead. A small quantity of aluminium (0.9% wt) and of zinc (0.15% wt) was detected in service pipes removed from Areas C and B respectively. Traces of copper were present in 7 of the 10 pipes. There was no apparent correlation between the presence of any particular trace metal and the occurrence of particulate or soluble lead.

Expert advice was sought from the Lead Development Association relating to the significance of these traces of metals for the corrosion of lead pipes, particularly in relation to the composition of the corrosion products formed. This source reported no known literature which had investigated such effects.

(c) X-ray diffraction - inorganic analysis of corrosion products

The theory of lead-water chemistry predicts that the corrosion deposits within a lead pipe will consist mainly of either or both normal lead carbonate ($Pb\ CO_3$) and basic lead carbonate ($Pb_3\ (CO_3)_2\ (OH)_2$). The carbonate type is determined predominantly by the pH and alkalinity of the water. The normal type occurs at low pH and the basic type at high pH; as alkalinity increases the pH for this stability transition increases. Under the conditions of alkalinity and pH existing for the sources, Table 5.5 basic lead carbonate is the predicted stable form. The X-ray diffraction studies have confirmed this prediction.

These results provide no consistent correlation between particulate lead problems and specific chemical components of the internal corrosion deposits. Basic lead carbonate was evident both at sites where particulate lead had occurred (e.g. Areas C and B), at sites previously demonstrating soluble lead problems (Area H) and in a non-problem area (Area I). However, sites in the last two categories were evidently experiencing minor levels of particulate lead during the current sampling programme.

Of note is the absence of lead-compounds in the material removed from the non-problem Area J pipe. This suggests that the internal lead-containing deposits are very well adhered to the surface. The material collected from the bore surface for the X-ray examination representing the upper layer of deposit is likely to have been amorphous and therefore undetectable by this technique. The SiO_2 detected may have arisen from contamination of the pipe interior during exhumation.

The additional analysis using X-ray fluorescence identified a number of other constituent elements in the 3 samples examined (Areas C and B). The presence of other metals confirms previous findings.

(d) Microbiological investigations

The degree of colonisation as determined by the plate count technique did not correlate with the observed levels of particulate lead in the water samples collected prior to service pipe exhumation or with the historical behaviour associated with each of the areas. It is, however, possible that because the bacterial matter is associated with deposits in the service pipes, the microbiological character of the pipe is not stable and can fluctuate with conditions in the pipe, e.g. retention time, flow rate, etc., as these themselves affect the deposits. The conditions preceding pipe exhumation may therefore be important. The pipe exhumed from an area supplied by ground water (Area I) supported much less microbiological activity.

6. DISCUSSION

The objectives of this work are to confirm the occurrence of particulate lead, to determine the circumstances under which particulate lead occurs, and to investigate remedial actions.

The national survey demonstrated the wide extent of the particulate lead problem within the UK Water Industry and successfully identified several locations where problems could be investigated. Using various investigative techniques it has been possible to obtain sufficient understanding of the particulate lead problem to suggest potential remedial measures.

An analysis of the initial data permitted a typical water chemistry with potential particulate lead problems to be defined. Particulate lead problems were found to be most prevalent in areas that are supplied by soft upland water with a low alkalinity, low hardness, often high colour and organics content. Often such supplies receive minimal treatment, resulting in high iron concentrations and occasionally high manganese and aluminium concentrations and poor pH control. This was supported by later trials associated with pipe exhumation where an area supplied by a spring source, typically of high hardness and alkalinity and of low TOC, was confirmed as a non-problem area.

The development of an analytical technique for speciation of different lead particle sizes allowed the identification of particulate lead problem areas. It also allowed the identification of factors that affect particulate lead by enabling a comparison of the size distribution of lead particles with other constituents of the water (such as iron, aluminium, manganese or TOC) to be conducted.

The occurrence of particulate lead has been confirmed in Areas A, B, C, D and L. Furthermore size fractionation analysis has revealed more than one type of particulate lead problem. These types have been called particulate/colloidal lead (typical of Areas C, E and F) and flaking lead (typical of Areas A and B). Area D seems to experience a combination of soluble and flaking lead.

The distinguishing feature between particulate/colloidal and flaking lead problems is size of the particulate matter. Flaking lead is characterised by particles >12µm in size sufficient to render them visible to the consumer. Particulate/colloidal lead is associated with particles of a size range of 0.08µm to 12µm and these are generally invisible to the consumer.

Sampling from the Area C pipe rig has shown that iron, together with organic matter, is associated with high lead levels, and removal of both iron and organic matter results in significant reductions in lead concentrations. Physical disturbance of the pipe rig also affected levels of particulate lead.

Work in the north of England has shown that average lead levels are reduced by phosphate dosing. The dosing also results in a reduction in average iron levels and it is thought that the reduction in iron is the most likely mechanism for the reduction in lead. However phosphate dosing does not eliminate all peaks in lead concentrations. The dosing of orthophosphate in Area D, where a combination of soluble and flaking lead occurs, has reduced lead levels. It is likely that this reduction is due largely to the reduction in soluble lead levels although this was not tested by size fractionation.

Sampling in Area A and B also showed that high lead levels are associated with high iron levels. Another factor which was shown to affect flaking lead here was physical disturbance of the deposits, i.e. fast flow, water hammer. This thus confirms previous findings.

In order to investigate further the apparent association of lead with iron and organics, lead corrosion layers were analysed. The corrosion products on the internal surface of lead service pipes are predominantly formed as two distinct layers, the bottom one probably a lead carbonate deposit attached firmly to the pipe surface. The upper corrosion layer can vary, particularly in iron, manganese and organic carbon concentrations and seems to control the differences in lead problems. Flaking and particulate lead problems in general appear to be associated with lead corrosion layers that are high in iron, manganese and in particular, organic carbon. Non-problem areas are associated with lead deposits rich in organic matter but no other metal. This supports the previous investigations with the Area C pipe rig where iron and organic material in combination are found in association with particulate lead. A limited investigation into a possible correlation between the appearance of particulate lead at the tap and microbiological activity, and specifically biofilms, provided no consistent evidence to link these two factors. Similarly variations in pipe metal composition, in particular with regard to contaminant types and level did not correlate with the occurrence of particulate lead.

The initial work has thus demonstrated that there is a strong association between iron and elevated levels of particulate lead. The removal of iron was considered a possible remedy and was therefore investigated during the later stages of the project.

One possible mechanism for high levels of particulate lead and iron at the tap was considered to be the entry of high levels of particulate iron into the lead service pipe which resulted in high levels of particulate lead either by adsorption or scouring. This was investigated using suitable filters between the service pipe and the distribution main. Although particulate iron was reduced by these filters, it was not entirely eliminated; furthermore particulate lead was not reduced. It thereby became clear that the association of particulate iron and particulate lead had already taken place in the form of an iron-lead corrosion layer on the inside of the service pipe. This could flake off periodically especially at times of rapid water flow resulting in particulate lead and iron at the tap, despite the presence of the filter. Thus a filter just upstream of the lead service pipe does not reduce particulate lead in the medium term (30 weeks). If the iron-lead corrosion layer forms as a result of particulate iron reaching the service pipe and then combining with the lead corrosion products, once the iron-lead layer is expended or removed it is conceivable that particulate lead would stop appearing at the tap, if a filter were in place, since particulate iron could be prevented from contacting the lead corrosion layer to replenish the reservoir. Alternatively the iron-rich layer may be accumulating as a result of soluble iron in the water. If this were the case, filters would have no effect and particulate lead would continue to appear.

The results of the filter experiments indicating an iron-lead corrosion layer as the origin of the particulate lead at the tap suggests that service pipe cleaning techniques should be investigated. A limited trial of service pipe cleaning took place during the later stages of the project when CO_2 scouring of two lead service pipes was conducted. The results were inconsistent. For one property, no statistically significant effect of CO_2 scouring on levels of particulate lead at the tap was recorded. However for the second property, CO_2 scouring eliminated particulate lead and caused a substantial drop in particulate iron concentrations. Particulate lead did not recur at the premises during the post-scour monitoring period (8 weeks). The results for two properties plus associated controls are considered insufficient from which to draw reliable conclusions. Continued investigations into the technique are recommended.

The objectives of this work have been met in the following ways. The occurrence of particulate lead has been confirmed at a number of locations in the UK. The circumstances under which particulate lead occurs have been identified as a particular water type i.e. soft upland source of a certain characteristic chemistry. An analytical method has been developed to identify the occurrence and type of particulate lead problems and this method has also been used to provide evidence for the association between high lead levels and high levels of iron and organic matter.

Field trials have confirmed that the occurrence of particulate lead is associated with high concentrations of particulate iron and organic carbon and is additionally promoted by physical disturbance of the service pipe. Remedial measures have been investigated and some of those examined appear promising. Depending on the circumstances, potential remedies are improvements in water treatment to reduce iron and organic carbon contents at source, if necessary combined with phosphate or silicate dosing to control iron in distribution. CO_2 scouring of lead service pipes may offer another potential remedy to problems of particulate lead.

7. CONCLUSIONS

(1) A national survey has shown that particulate lead problems are experienced throughout the UK Water Industry and are most prevalent in areas that are supplied by soft upland water with a low alkalinity, low hardness, often high colour and organics content. Often such supplies receive minimal treatment, resulting in high iron concentrations and occasionally high manganese and aluminium concentrations and poor pH control.

(2) Two types of particulate lead problems have been identified. Flaking lead is characterised by particulate lead >12µm in size and thereby rendered visible to the consumer. Particulate/colloidal lead is associated with particulate lead between 0.08 and 12µm in size which is invisible to the consumer.

(3) Size fractionation is a useful method for establishing the presence and the type of particulate lead.

(4) Elevated levels of particulate lead in Area C are caused by high levels of iron and organic matter in the treated water. Removal of the iron and organic matter is an appropriate solution to the problem of particulate lead in Area C.

(5) In Areas A,B and L, particulate lead appears to be associated with elevated iron concentrations. Treatments, such as phosphate dosing, that reduce average iron levels also successfully reduce average lead levels. Mechanical disturbance of the service pipes such as fast flow conditions and water hammer leads to an increase in particulate lead in the tap water.

(6) Corrosion layer analysis has shown an association between iron and organic carbon with lead in the upper corrosion layer in lead service pipes. This confirms the results of water analyses which show a similar association in some areas.

(7) Filtering experiments in Area B have indicated that 1µm pore filters just upstream of the lead service pipe reduce but do not eliminate particulate iron at the tap. Levels of particulate lead are unaffected. There is believed to be a reservoir of particulate iron within the service pipe which serves to initiate particulate lead problems. Filters therefore are not effective at reducing lead levels in the medium term (up to 30 weeks).

(8) CO_2 scouring eliminated particulate lead at one property in the short term. At a second property no statistically significant effect was recorded. Due to the limited geographic extent of sampling, reliable conclusions on the effectiveness of CO_2 scouring as a means of reducing or eliminating particulate lead can not be given from the data generated to date.

(9) Limited investigations into a possible correlation between the appearance of particulate lead at the tap and the presence of microbiological activity and specifically biofilms, provided no consistent evidence to link these two factors.

(10) Variations in pipe metal composition, in particular with respect to contaminant types and levels, did not correlate with the occurrence of particulate lead.

8. RECOMMENDATIONS

(1) The improved treatment at the water treatment works supplying Area C to remove iron and organic matter should result in reduced lead levels in distribution. Regular monitoring of lead levels in distribution is recommended. (It is suggested that silicate or polyphosphate dosing is performed on a trial basis.)

(2) In areas with severe particulate iron and lead problems polyphosphate or orthophosphate dosing is recommended as a short term solution, initially on a trial basis. These techniques may reduce iron levels and thereby may possibly have an effect on lead levels. It is likely that the effects of this dosing may be noticeable only in the long term since a reservoir of iron-rich corrosion deposit will remain in the lead service pipe for some time. It is likely that these solutions will not enable 100% compliance with the PCV and lead pipe replacements may be the only long-term solution.

(3) A combination of, where appropriate, improvements in water treatment to reduce iron and organic carbon; chemical dosing or mains rehabilitation to reduce iron in distribution; mains cleaning to remove the reservoir of particulate iron in the distribution system, is recommended as a medium term solution to reduce particulate lead problems throughout the UK. Individual circumstances pertaining in the problem area will determine which combination of remedial techniques will be needed.

(4) More comprehensive long-term trials to investigate the effectiveness of CO_2 cleaning lead service pipes are recommended.

There is no doubt that the eventual solution to problems of elevated lead levels will be pipe replacement. In the interim period the remedies listed above are recommended.

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APPENDIX A

SIZE FRACTIONATION STUDIES: METHOD DEVELOPMENT

Adapted from:

SIZE FRACTIONATION STUDIES

Susan J Cowling and A M Gunn (WRc Medmenham)

A.1 Background

Lead can exist in natural waters in a great variety of physicochemical forms, including free aquo ions, inorganic ion-pairs and complexes, organic complexes, associations with highly dispersed colloidal material, as suspended particles of insoluble salts, or adsorbed on inorganic particulates. These various species can be classified on the basis of size distribution, as shown in Figure A.1, and size fractionation procedures can therefore prove extremely useful in investigations of lead speciation. Harrison and co-workers (Laxen and Harrison 1981, de Mora et al. 1987) have described a scheme involving filtration to five different size fractions coupled with measurements of 'labile' lead as defined by anodic stripping voltammetry and Chelex chelating ion-exchange resin techniques. This basic approach was considered appropriate for the present study. This involved filtration of the tapwaters through Nuclepore polycarbonate filters of pore sizes 12, 1, 0.4, 0.08 and 0.015µm and determination of lead, iron, manganese, aluminium and total organic carbon (TOC) in each fraction. Polycarbonate filters were preferred to cellulose acetate or nitrate membranes because they provide better size discrimination at small pore sizes (Laxen and Chandler 1982) and are less prone to adsorption problems (Gardner 1982).

APPROX. SIZE (nm dia, log scale)	10000		100	10	1	
CLASS	Minerals and precipitates	Adsorbed on inorganic particulates	Highly dispersed colloidal material	Bound to macromolecules	Organic chelates, other inorganic ions, ion pairs and complexes	Free aquo ions
EXAMPLES	$PbCO_3(s)$	Adsorbed on hydrous iron and manganese oxides and clay minerals	Adsorbed on hydrous iron and manganese oxide colloids	Pb fulvic acid complexes	Pb-EDTA $PbCO_3$	Pb^{2+}

Figure A.1 - Classification scheme for lead species in water (adapted from Fielding and Hunt 1984)

A.2 Adsorption during filtration

Loss of lead to the filter unit by adsorption can be a serious problem in size fractionation schemes, particularly for samples of low hardness and high pH, as encountered in this work. In order to minimise such effects, filter systems were preconditioned with 0.1M calcium nitrate, then the sample itself.

The larger pore sizes (12, 1 and 0.4µm) were used in Sartorius 250ml, 47mm polycarbonate filter units. The combination of this unit and the 0.4µm Nuclepore filters has previously been tested for lead adsorption losses at WRc by Gardner (1982). Synthetic samples prepared to contain 100mg $NaNO_3$/l as a supporting electrolyte and to have lead concentrations of around 20µg/l showed losses of between 10 and 20% across the pH range 6 to 9. Given the absence of any competing cations other than sodium, these results can be taken to be indicative of a 'worst possible case' for adsorption and the procedure was therefore considered adequate for the present purposes.

The smaller pore sizes (0.08 and 0.015µm) were used in an Amicon 50ml, 43mm stirred ultrafiltration cell (Model 8050) which could be operated at a much higher pressure and provided faster sample flow rates. Initial adsorption tests on this unit were undertaken using 0.08µm pore size filters. A prefiltered low hardness artificial tapwater (10mg Ca/l, 1.6mg Mg/l, 7mg Na/l, 0.8mg K/l) was spiked with 100µg Pb/l and pH adjusted, then three consecutive 50ml aliquots were run through the same 0.08µm filter. Lead concentrations were determined before and after filtration and the losses calculated (see Table A.1). The test was performed four times at pH 6.5 and three times at pH 8.

Table A.1 - Results of adsorption tests on 0.08µm filters

	PERCENTAGE LOSS OF LEAD	
	At pH 6.5	At pH 8
1st aliquot	18 ± 6	28 ± 7
2nd aliquot	6 ± 6	19 ± 7
3rd aliquot	2 ± 4	23 ± 7

Confidence intervals are at the 95% level.

Later tests on the 0.015µm filters have indicated that much greater losses may occur when filtration times are very slow (> 1 hour) and the pH is high (>pH 8).

A.3 Procedure

Samples are collected in acid-washed two-litre polyethylene bottles and returned to the laboratory where all filtrations are completed within 48 hours.

The filtrations for 12, 1 and 0.4µm are performed in parallel using acid washed Sartorius polycarbonate units. Preparation of the units is achieved by filtering 100ml of 5% nitric acid, followed by 150ml of deionised water, 250ml of 0.1M calcium nitrate, 250ml of deionised water then 50ml of the sample. Then 250ml of the sample is filtered, of which 60ml is stored at 4°C in borosilicate glass bottles for TOC analysis and the remainder preserved with 5ml/l Aristar nitric acid and stored in polyethylene bottles for metals analysis.

Prior to undertaking filtrations to 0.08 and 0.015µm in the Amicon 8050 ultrafiltration unit, samples are prefiltered to 0.4µm to reduce the likelihood of clogging. The system is prepared by rinsing with 50ml of 0.5% v/v nitric acid and filtering a small amount of the same, then repeating this procedure with, in turn, deionised water, 0.1M calcium nitrate, deionised water again, and finally the sample itself. As these filtrations can be very slow, only 20ml is taken for TOC analysis and 20ml for metals analysis. The samples are preserved as above.

A.4 References

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APPENDIX B

REPORT ON INVESTIGATIONS INTO THE OCCURRENCE OF PARTICULATE LEAD IN AREA A

APPENDIX B

REPORT ON INVESTIGATIONS INTO THE OCCURRENCE OF PARTICULATE LEAD IN AREA A

B.1 Area and Source Details

Area A has a population of ~50,000 consumers served by lead service pipes. There are 3 supplies. The major supply, Source 1 is a moderately soft moorland water with historical colour problems and high aluminium residuals because of inadequate treatment. Source 2 is river derived. Aluminium, iron and manganese only occasionally exceed the PCV. Source 3 is river water filtered through natural gravel beds before abstraction. Iron, aluminium and manganese are well below the PCV. These supplies are mixed depending on demand and availability. All these sources are soft, have a low alkalinity and a relatively high pH. The water is not plumbosolvent.

Area A suffers from a flaking lead problem, where lead is present in the form of "tea leaves" and visible to the consumer. Flaking lead is a seasonal problem with highest occurrence in the summer period. The problem does not occur every summer and it does not always recur in all premises with lead service pipes. The problem is decreasing in size as lead service pipes are being replaced. The flaking lead problem has been investigated since 1984 as a joint exercise between the Water Utility and WRc.

B.2 Surveys in 1984

In 1984, 112 random daytime tap samples were taken from a small part of Area A and analysed for lead, aluminium, iron, manganese, copper and zinc.

More than 21% of the samples taken from the kitchen tap exceeded the then EC MAC for lead of 100µg/l. These high lead values were nearly always associated with iron concentrations >100µg/l, see Table B.1.

Table B.1 - Number and percentage of samples with iron concentrations exceeding 100µg/l in various categories of lead concentrations - 1984 survey

LEAD CONCENTRATION (µg/l)	SAMPLES EXCEEDING 100µg/l IRON	
	No.	%
≤ 20	1 of 44	2.3
21 - 50	1 of 32	3.1
51 - 100	2 of 11	18.2
101 - 200	9 of 11	81.8
>200	12 of 13	92.3

The distribution of iron concentrations is given in Table B.2

Table B.2 - Distribution of iron concentrations - 1984 survey

IRON CONCENTRATION (µg/l)	% OF SAMPLES
≤ 50	48.4
51 - 100	30.3
101 - 200	8.9
201 - 500	7.1
>500	5.3

Very high lead concentrations were always associated with high iron, aluminium and manganese levels. The ratio lead:iron was approximately 1.5 in samples where lead was >100µg/l. No high iron, aluminium or manganese concentrations were found in premises with low lead levels. In addition to the kitchen tap samples some premises were also sampled at the bathroom tap. There are not sufficient data on bathroom samples to allow a proper analysis, but all properties sampled that gave high bathroom results (lead > 100µg/l) showed low kitchen tap values for lead. This indicates that the extent of the flaking lead problem was larger than 21% of the properties sampled as indicated by the kitchen tap survey. Hydrant samples were taken from mains directly supplying the problem houses. In these samples no lead was found, but high concentrations of iron, aluminium and manganese were present. Particulate matter in the problem houses consisted of large black particles and fine brown material. Both types of particles had the same composition: lead, iron and sometimes manganese but they did not contain aluminium.

B.3 Surveys in 1986

In 1986 samples were taken prior to and following remedial action in the distribution system. In one zone mains cleaning was conducted and in another zone orthophosphate was dosed; a third zone was used as a control zone. In none of the zones were high lead concentrations found. As a result, it was not possible to identify any beneficial effect of remedial action.

B.4 Surveys in 1988

B.4.1 Large scale survey

After a very mild winter (1987 - 1988), complaints of black bits in the water recurred. In contrast to 1984 complaints started early, in mid-February. The area affected was approximately 1 square mile.

In April 1988, 190 premises were sampled from inside and outside the problem area, all supplied by the same type of water. 75% of samples were taken from lead-plumbed houses and 25% from premises with another or unknown type of pipe material.

Samples were taken as random daytime samples and after flushing the service pipe. In random daytime samples 3% exceeded the then MAC for lead (100 µg/litre), in the samples taken after flushing 6% were in excess of this MAC. Table B.3 shows that there was no significant and no consistent difference between lead, iron and aluminium concentrations in random daytime and flushed samples. But in samples where flaking lead is present, flushed samples tend to show higher lead, iron and aluminium concentrations (see Table B.4)

Table B.3 - Comparison of random daytime (rdt) and flushed samples - 1988 survey

SAMPLE	LEAD % OF SAMPLES	IRON % OF SAMPLES	ALUMINIUM % OF SAMPLES
Flushed > rdt	11.2	18.2	41.2
Flushed < rdt	21.9	12.3	53.5
Flushed = rdt	66.9	69.5	5.3

Table B.4 - Comparison of random daytime (rdt) and flushed samples with lead concentrations exceeding 100µg/l - 1988 survey

SAMPLE	LEAD % OF SAMPLES	IRON % OF SAMPLES	ALUMINIUM % OF SAMPLES
Flushed > rdt	66.7	75.0	83.3
Flushed < rdt	33.3	25.0	16.7
Flushed = rdt	0	0	0

The premises with high lead concentrations in the water also showed an increased level of iron and aluminium. The ratio between lead and iron varied between 1.1 and 2.5 and was on average 1.5. Increased aluminium concentrations were also found in some premises where lead was absent (Pb <20µg/l, see Table B.5). High iron was rarely found in premises where lead was absent, Table B.5.

Table B.5 - Number and percentage of samples with iron and aluminium concentrations exceeding 100µg/l in various categories of lead concentrations - 1988 survey

LEAD CONCENTRATION (µg/l)	SAMPLES EXCEEDING 100µg/l IRON		SAMPLES EXCEEDING 100µg/l ALUMINIUM	
	No.	%	No.	%
≤ 20	8 of 161	5.0	36 of 161	22.4
21 - 50	2 of 11	18.2	1 of 11	9.1
51 - 100	0 of 6	0	3 of 6	50.0
101 - 200	1 of 1	100	1 of 1	100
> 200	11 of 11	100	7 of 11	63.6

The distribution of iron and aluminium concentrations is given in Table B.6.

Table B.6 - Distribution of iron and aluminium concentrations - 1988 survey

IRON		ALUMINIUM	
µg/l	% OF SAMPLES	µg/l	% OF SAMPLES
≤ 50	79.5	≤ 50	0
51 - 100	8.9	51 - 100	74.7
101 - 200	4.2	> 100	25.3
201 - 500	3.7		
> 500	3.7		

Increased manganese concentrations were only rarely found in premises where flakes were present. The particles found were both large black bits and small amorphous brown bits. Hydrant samples from mains supplying the area indicated that the water in the area was clean. A total water quality analysis in a small number of houses indicated that at the time of sampling water quality within the problem area was not significantly different from the water quality in a non-problem area.

Table B.7 lists the full range of parameters monitored.

B.4.2 Detailed '6 premises' survey

A comprehensive sampling exercise was undertaken at 3 premises in the problem area and 3 in the non-problem area with the following objectives:

(a) to determine under which circumstances, hydraulic and chemical, the lead corrosion layer becomes destabilised and becomes detached thus resulting in particulate lead,

(b) to identify any significant differences in water quality in the problem and non-problem area,

(c) to identify the source of the particles (mains or service pipes), and

(d) to determine if any association exists between lead, iron, manganese and aluminium using size fractionation studies.

The sampling procedure outlined in Table B.8 was adopted to provide suitable samples.

The chemical analysis of the water from premises in the problem and non-problem areas showed no significant difference in water quality at the times the samples were taken (see Table B.9). In one premises colour and turbidity were higher than in the others but this is likely to be a result of high concentrations of metal in the water, and not the cause.

Table B.10 presents the results of samples taken under various circumstances as detailed in Table B.8.

Table B.7 - Total water analysis from 16 properties - 1988 survey

PREMISES IDENTI-FICATION	TYPE OF SERVICE	pH	CONDU-CTIVI-TY µS/cm 20°C	TURBI-DITY NTU	COLOUR Hazen	TOC mg C/l	FREE CHLOR-INE mg/l	ALKAL-INITY mg/l $CaCO_3$	Na mg/l	K mg/l	Cu µg/l	Mg mg/l	Ca mg/l	Al µg/l	Pb µg/l	Mn µg/l	Fe µg/l
1	Lead	7.9	115	0.8	<2	0.97	0.15	22	10.4	0.9	10	2.2	11.2	78	8	5	20
2	No lead	8.0	123	0.7	<3	0.96	0.10	31	9.2	1.1	19	2.5	13.7	73	8	5	43
3	No lead	8.1	141	0.5	<2	0.93		39	8.0	1.3	20	2.8	16.0	69	10	8	66
4	Lead	8.0	139	0.3	<2	0.96		39	8.7	1.4	19	3.0	16.1	56	11	8	20
5	Unknown	8.1	140	0.5	<2	0.70	0.50	40	8.9	1.2	<5	3.2	16.9	56	11	9	29
6	No lead	8.3	138	0.3	<2	0.86	0.50	40	8.8	1.3	<5	3.1	16.5	51	8	5	<20
7	No lead	8.2	138	0.3	<2	0.91	0.30	40	8.2	1.2	17	2.9	15.7	59	9	<5	<20
8	Lead	8.1	141	0.4	<2	0.82	0.40	39	8.2	1.0	<5	3.0	15.8	72	46	9	40
9	Lead	8.0	102	0.2	<2	0.84	0.10	12	11.7	0.7	9	1.6	7.8	113	8	<5	42
10	Lead	8.1	125	0.5	2	0.92	0.10	31	8.4	1.0	127	2.3	13.1	563	7100	148	2820
11	Lead	7.7	115	0.2	<2	0.84		22	9.5	0.8	12	1.9	10.3	76	14	10	22
12	No lead	7.9	117	0.3	<2	1.10		23	9.9	0.9	13	2.0	10.5	78	<8	8	<20
13	No lead	7.7	102	1.1	2	0.88	0.10	11	11.2	0.4	6	1.6	7.6	110	<8	12	45
14	Lead	7.7	102	0.4	<2	0.82	0.10	11	11.0	0.4	<5	1.6	7.6	145	29	45	76
15	Lead	8.1	128	0.4	2	0.83	0.10	31	9.6	1.1	35	2.6	14.5	89	18	40	111
16	No lead	8.1	143	0.4	<2	1.0	0.50	39	8.9	1.3	6	3.2	17.0	61	<8	8	34
		8.0	105	0.2	2	0.79	0.10	12	11.3	0.3	<5	1.6	7.7	103	<5	<5	109

Table B.8 - Sampling programme for '6 premises' survey

STEP	PROCEDURE
1	Flush pipes for 2 minutes measure pH, temperature, residual chlorine, dissolved oxygen (DO).
2	Take sample with high flow (sample A) - 1 litre. Filtration in laboratory 12 and 0.4µm. Analysis: Fe, Al, Mn, Pb, TOC.
3	Stagnate water for 30 minutes. Take sample with normal flow (sample B) - 50ml. Take sample with normal flow and filter over 0.40µm at premises (sample C). Analysis: Fe, Al, Mn and Pb.
4	Sample with normal flow (sample D) - 2 litre. Filtration in laboratory (12, 1.0, 0.04, 0.08, 0.015µm). Analysis: Fe, Al, Mn, Pb, and TOC.
5	Sample with fastest flow and water hammer (sample E) - 1 litre. Filtration in laboratory 12 and 0.4µm. Analysis: Fe, Al, Mn, Pb and TOC.

Table B.9 - Chemical analysis of tap water samples in problem area and in non-problem area

AREA	pH	COLOUR Hazen	DISSOLVED O_2 %	TOC mg C/l	FREE CHLORINE mg/l	ALKALINITY mg/l $CaCO_3$	TURBIDITY NTU	Na mg/l	K mg/l	Cu mg/l	Mg mg/l	Ca mg/l	Pb µg/l	Fe µg/l	Mn µg/l	Al µg/l
Non-problem 1	8.1	3	86	0.9	0.10	13	0.6	11	0.6	0.02	1.4	7.4	27	108	13	118
Non-problem 2	7.8	2	98	0.9	0.20	33	0.4	9	1.0	0.03	2.5	13.3	12	30	6	67
Non-problem 3	7.9	<2	87	0.8	0.40	41	1.0	8	1.2	0.006	3.0	15.7	21	56	23	56
Lead problem 1	7.8	4	95	0.9	0.15	33	1.8	8	1.0	<0.005	2.5	13.1	203	184	33	91
Lead problem 2	7.8	2	100	0.9	0.25	34	0.5	9	1.0	<0.005	2.4	13.0	41	31	6	61
Lead problem 3	7.8	<2	99	0.9	0.25	33	0.3	9	1.0	<0.005	2.5	13.6	13	34	6	71

Table B.10 - Effect of hydraulic circumstances on total metal concentrations in tap water

		LEAD (µg/l)				IRON (µg/l)				ALUMINIUM (µg/l)				MANGANESE (µg/l)				TOC (mg C/l)			
		A	B	D	E	A	B	D	E	A	B	D	E	A	B	D	E	A	B	D	E
Non-problem houses	6	5	23	20	28	56	56	67	81	98	92	99	110	7	5	8	11	1.08	-	1.44	1.43
	5	8	20	27	12	34	38	34	40	60	63	61	61	5	4	5	5	2.68	-	1.24	1.58
	1	12	18	43	26	23	22	30	71	45	40	49	71	5	4	5	26	1.48	-	1.22	1.02
Problem houses	2	488	125	194	481	448	127	173	403	145	81	92	146	62	14	32	82	1.31	-	1.24	1.26
	3	96	133	51	168	50	66	38	52	62	92	58	71	12	8	9	10	1.31	-	2.24	1.05
	4	15	48	33	487	65	39	38	439	59	60	58	147	27	7	6	89	1.36	-	1.31	1.04

Sample A - 1 litre sample after flushing with high flow;
Sample B - 50ml sample after 30 minutes stagnation, normal flow;
Sample D - 2 litre sample after 30 minutes stagnation, normal flow;
Sample E - 1 litre sample, fast flow and water hammer.

Not much variation is found in non-problem houses as a result of different hydraulic circumstances. In problem houses there is a tendency for concentrations of lead, iron, aluminium and manganese to be higher in samples taken with high flow, and high flow plus water hammer (samples A and E). Stagnation in the lead pipes does not significantly increase metal concentrations (samples B and D). The lower sampling velocity of these samples may however have influenced metals levels. TOC concentrations do not vary significantly or consistently.

The percentage of particulate material (lead, iron and aluminium) >12µm and >0.40µm is higher in houses with lead problems compared to non-problem houses (Table B.11).

In non-problem houses the percentage of particulate material present does not differ significantly with hydraulic circumstances.

In problem houses the percentage of big particles >12µm tends to increase at high velocity and water hammer (samples A and E, Table B.11) compared to samples taken after stagnation with a normal flow (sample D, Table B.11). This is the case for lead, aluminium and iron.

No such effect was found in problem houses on the percentage of particles >0.40µm.

Thus adverse hydraulic circumstances (high flow and water hammer) causes an increase in the percentage of large particles (lead, iron and aluminium) present in tap water.

Size fractionation is a method used to distinguish between various size classes of material present in the water. Filtration over 0.40µm is the normally used method to distinguish between soluble and particulate material and often results in an under-estimation of particulate and colloidal material present. Size fractionation provides a much greater degree of discrimination.

The distinction between soluble and colloidal is somewhere around 0.015µm. In size fractionation, 5 different types of filters are used: 12, 1, 0.4, 0.08 and 0.015µm. Care is required in the interpretation of the 0.015µm cut-off as a significant quantity of lead may adsorb onto these fine filters. To calculate percentage of particulate material present the 0.08µm cut-off was used.

Results of size fractionation analysis are summarised in Table B.12. These show that a higher percentage of metal is present as particles than might be concluded from the customary method (filtration over 0.40µm).

Table B.11 - Percentage of particles >12µm and >0.4µm present in various samples in non-problem and in problem houses

TYPE OF PREMISES		AFTER FLUSHING HIGH FLOW SAMPLE A		AFTER 30 MINUTES STAGNATION NORMAL FLOW SAMPLE D		HIGHEST FLOW AND WATER HAMMER SAMPLE E	
		% >12µm	% >0.4µm	% >12µm	% >0.4µm	% >12µm	% >0.4µm
LEAD							
Non-problem:	6	0	17	16	47	16	41
	5	15	34	13	36	2	35
	1	7	26	5	29	16	73
Problem:	2	77	77	72	84	69	85
	3	38	54	19	36	55	65
	4	8	68	6	26	67	70
IRON							
Non-problem:	6	0	23	16	46	24	43
	5	0	38	0	34	16	37
	1	12	48	4	54	15	78
Problem:	2	79	79	66	86	69	86
	3	19	54	1	39	19	47
	4	11	68	3	26	60	88
ALUMINIUM							
Non-problem:	6	0	10	0	12	6	17
	5	0	5	0	5	0	7
	1	1	21	11	13	27	46
Problem:	2	50	50	23	32	37	41
	3	0	18	0	8	4	19
	4	8	21	0	9	40	47

Table B.12 - Results of size fractionation analysis - percentage of lead, iron and aluminium in each size class

PREMISES IDENTITY	TYPE OF PREMISES	% OF SAMPLE				LEAD	IRON	ALUMINIUM
		LEAD	IRON	ALUMINIUM				
	NON-PROBLEM:							
6	>12μm	16	16	0	Total conc.	20μg/l	67μg/l	99μg/l
	1 - 12μm	18	24	8	>0.4μm	48%	46%	12%
	0.4 - 1μm	14	6	4	>0.08μm	92%	90%	20%
	0.08 - 0.4μm	45	44	7				
	0.015 - 0.08μm	8	10	26				
	<0.015μm	0	0	54				
5	>12μm	13	0	0	Total conc.	27μg/l	34μg/l	61μg/l
	1 - 12μm	7	16	4	>0.4μm	36%	34%	5%
	0.4 - 1μm	16	18	1	>0.08μm	71%	34%	17%
	0.08 - 0.4μm	35	0	12				
	0.015 - 0.08μm	22	55	16				
	<0.015μm	7	11	67				
1	>12μm	5	4	11	Total conc.	43μg/l	30μg/l	49μg/l
	1 - 12μm	15	36	0	>0.4μm	28%	54%	13%
	0.4 - 1μm	8	14	2	>0.08μm	55%	81%	14%
	0.08 - 0.4μm	26	25	0				
	0.015 - 0.08μm	24	13	10				
	<0.015μm	21	6	76				
	PROBLEM:							
2	>12μm	72	66	23	Total conc.	194μg/l	173μg/l	92μg/l
	1 - 12μm	11	15	0	>0.4μm	84%	86%	32%
	0.4 - 1μm	1	5	9	>0.08μm	96%	86%	41%
	0.08 - 0.4μm	10	0	9				
	0.015 - 0.08μm	2	10	14				
	<0.015μm	2	4	45				
3	>12μm	19	1	0	Total conc.	51μg/l	38μg/l	58μg/l
	1 - 12μm	10	32	1	>0.4μm	35%	39%	8%
	0.4 - 1μm	6	6	7	>0.08μm	65%	83%	21%
	0.08 - 0.4μm	28	44	13				
	0.015 - 0.08μm	29	16	24				
	<0.015μm	6	1	55				
4	>12μm	6	3	0	Total conc.	33μg/l	38μg/l	58μg/l
	1 - 12μm	12	6	1	>0.4μm	27%	26%	9%
	0.4 - 1μm	9	17	8	>0.08μm	60%	27%	15%
	0.08 - 0.4μm	33	0	5				
	0.015 - 0.08μm	32	59	1				
	<0.015μm	8	14	84				

An important part of the lead and iron is present as particulate material, the percentage of particulate aluminium is relatively much less. This indicates that a major part of the aluminium present in the samples is in soluble or small colloidal form.

Where lead concentrations are high (premises No. 2) most iron and lead and a large part of the aluminium are present in the size class >12µm, indicating that these metals are present in the same, large particles. Unfortunately, this was the only size fractionation sample with an overall high metal concentration.

Detailed analytical results for all these samples are given in Table B.13 to B.16.

Table B.13 - Sample A - Fast flow without stagnation

PREMISES IDENTITY	SIZE FRACTION *	METAL CONCENTRATIONS (µg/l)				TOC
		LEAD	ALUMINIUM	MANGANESE	IRON	(mg C/l)
1	T	11.7	45.0	4.59	23.3	1.48
	12	10.9	44.5	3.87	20.5	-
	0.4	8.6	35.4	1.89	12.1	1.27
2	T	488.0	145.0	62.2	448.0	-
	12	111.0	72.5	17.0	95.5	1.31
	0.4	113.0	71.9	13.5	93.4	1.62
3	T	96.3	61.9	12.0	50.0	1.31
	12	59.8	63.7	8.51	40.3	1.44
	0.4	44.5	50.8	2.77	23.0	-
4	T	15.5	59.3	27.3	64.6	1.36
	12	14.2	54.7	20.2	57.6	1.41
	0.4	4.9	46.6	6.28	20.9	1.22
5	T	8.5	60.5	4.80	33.9	2.68
	12	7.2	60.8	4.57	34.6	1.16
	0.4	5.6	57.5	1.92	21.1	1.13
6	T	5.2	97.6	7.34	55.9	1.08
	12	5.2	97.2	4.12	58.0	1.30
	0.4	4.3	87.8	2.39	42.8	1.16

* T = Total concentration
12 = <12µm fraction
0.4 = <0.4µm fraction

Table B.14 - Sample B - Normal flow, unfiltered and filtered to 0.4µm, on site, 30 minutes stagnation

SAMPLE *	METAL CONCENTRATIONS (µg/l)			
	LEAD	ALUMINIUM	MANGANESE	IRON
1	18.2	40.0	4.29	22.5
1f	16.0	43.5	2.77	17.3
2	125.0	80.7	14.3	127.0
2f	89.6	72.7	10.6	72.9
3	133.0	91.7	8.41	66.5
3f	83.2	56.6	2.11	26.0
4	48.1	59.7	6.77	38.7
4f	39.4	57.5	4.27	33.9
5	19.8	63.0	4.32	38.4
5f	23.0	58.8	2.75	24.0
6	22.9	92.2	5.33	55.6
6f	9.8	89.2	2.57	47.4

* f = filtered

Table B.15 - Sample D - Size fractionation

PREMISES IDENTITY	SIZE FRACTION *	METAL CONCENTRATIONS (µg/l)				TOC
		LEAD	ALUMINIUM	MANGANESE	IRON	(mg C/l)
1	T	43.4	48.7	5.16	30.4	1.22
	12	41.0	43.3	4.88	29.2	-
	1	34.4	41.4	2.45	18.1	-
	0.4	30.9	42.1	1.50	13.8	1.06
	0.08	19.6	36.8	1.35	6.03	1.14
	0.015	9.1	37.1	-	1.95	1.81
2	T	194.0	91.9	31.9	173.0	1.24
	12	54.4	70.7	10.1	58.3	-
	1	32.2	58.7	6.01	32.6	-
	0.4	30.2	62.6	5.84	23.6	-
	0.08	9.9	54.1	2.80	5.76	1.96
	0.015	5.0	41.6	1.37	6.39	-
3	T	51.2	57.9	8.73	38.0	2.24
	12	41.4	55.3	6.41	37.7	-
	1	36.0	57.0	2.75	25.4	-
	0.4	32.7	53.0	2.04	23.2	1.78
	0.08	18.2	45.5	1.64	6.40	2.89
	0.015	3.3	31.7	1.36	-0.25	1.40
4	T	32.6	57.8	6.07	38.3	1.31
	12	30.7	61.2	5.24	37.0	-
	1	26.8	57.1	2.45	34.7	-
	0.4	24.0	52.5	1.49	28.0	1.51
	0.08	13.2	49.3	1.56	4.29	1.29
	0.015	2.7	48.7	1.88	5.44	0.95
5	T	27.3	61.4	4.76	34.1	1.24
	12	23.7	63.2	4.56	34.1	-
	1	21.8	58.8	2.33	28.7	-
	0.4	17.4	58.2	1.47	22.6	1.56
	0.08	7.8	50.6	0.81	3.57	1.89
	0.015	1.8	40.9	1.57	3.91	0.90
6	T	19.6	99.0	7.94	67.2	1.44
	12	16.5	106.0	5.34	56.1	-
	1	13.0	90.5	2.66	40.2	-
	0.4	10.3	86.6	2.06	36.3	2.12
	0.08	1.5	79.5	1.80	6.85	1.12
	0.015	0.0	54.0	2.62	-0.50	1.23

* T = Total concentration, other fractions in µm (figure defining upper limit)

Table B.16 - Sample E - Water hammer

PREMISES IDENTITY	SIZE FRACTION *	METAL CONCENTRATIONS (µg/l)				TOC
		LEAD	ALUMINIUM	MANGANESE	IRON	(mg C/l)
1	T	26.3	71.0	25.7	70.6	1.02
	12	22.1	52.0	18.8	60.0	1.12
	0.4	7.1	38.0	3.89	15.6	-
2	T	481.0	146.0	81.8	403.0	1.26
	12	150.0	91.3	19.8	123.0	0.86
	0.4	70.8	86.5	10.9	54.5	0.92
3	T	168.0	71.1	9.82	51.9	1.05
	12	75.3	68.2	7.26	41.9	1.86
	0.4	58.5	57.8	2.98	27.4	0.83
4	T	487.0	147.0	89.3	439.0	1.04
	12	161.0	88.7	44.7	177.0	1.36
	0.4	145.0	77.9	34.7	51.6	1.10
5	T	12.3	60.9	4.99	39.8	1.58
	12	12.1	62.6	4.52	33.2	0.84
	0.4	8.0	56.3	2.12	25.2	1.13
6	T	28.0	110.0	1.5	81.1	1.43
	12	23.4	103.0	7.21	61.9	1.13
	0.4	16.5	94.5	3.63	46.0	0.85

* T = Total concentration
12 = <12µm fraction
0.4 = <0.4µm fraction

Consumer questionnaires were also undertaken at the 6 premises. From the 3 consumers interviewed outside the problem area, one mentioned discoloured water problems in December and January. Problems were associated with works on the mains and disappeared quickly. This complainant does live outside of, yet close to, the problem area.

All 3 consumers interviewed in the problem area mentioned dirty water with black bits. Some mentioned the chlorination and others sewerage renovation works going on in the vicinity.

This information was obtained from a small number of consumers and thus is of limited value.

The conclusions of this detailed study are as follows. The occurrence of high lead concentrations coincides with high iron and aluminium and sometimes manganese. No relationship was found with TOC or any other water quality parameter. The problem is concentrated in a small area. It is not always possible to reproduce high metal concentrations.

There are indications that non-lead plumbed houses do not show increased metal levels. In the non-problem area hydraulic circumstances have no significant effect on water quality. In the problem houses flushing and water hammer both tend to lead to an increase in lead, iron, aluminium and manganese. Stagnation in lead pipes does not significantly alter metal concentrations. Only one of the size fractionation samples showed high lead levels. Most of this lead was present as large particles (>12µm) probably associated with iron, aluminium and manganese.

In all samples a high percentage of the metals is present in particulate form, except aluminium that is mostly present in solution.

B.5 Surveys in 1989

B.5.1 Background

In 1989 a stability survey was carried out. Fortnightly samples were taken at selected premises (slow flow and fast flow) from March till October. Samples were analysed for lead and iron and a total water analysis was carried out on all fast flow samples. The presence and absence of flakes was judged visually. The objective of this exercise was to monitor water quality at selected points in Area A both before and during the expected increase in occurrence of flakes. The monitoring would indicate the start of the problem period when a detailed sampling exercise at a larger number of premises was planned. This detailed survey in which 240 premises were sampled was carried out in June 1989. Unfortunately the flaking lead problem did not recur (or only on a very limited scale) in the summer of 1989.

B.5.2 Stability survey

In total 8 premises (2 non-lead plumbed and 6 lead plumbed) were sampled on a 2-weekly basis. In none of the slow flow samples were lead concentrations in excess of 100µg/l found. In 2 lead-plumbed premises, flakes were occasionally reported and lead concentrations in excess of 100µg/l were measured. Compared to other years, lead concentrations were relatively low. Also iron and aluminium concentrations were low compared to problem years. In the 2 premises that experienced some flakes in the water no obvious link with changes in water quality could be found. Because of the absence of an obvious flaking lead problem, no further detailed analysis of the data has been carried out.

B.5.3 Special survey

In June and July 1989, 240 premises from all over Area A were sampled. Two types of samples were taken, a slow flow sample and a fast flow sample. In 14 out of 190 premises (7.4%) the first sample taken with a slow flow exceeded the then MAC for lead. In 6 premises (3.2%) the then MAC was exceeded in the sample taken with a fast flow. The survey showed that there was no major occurrence of flakes this year. This results in relatively low lead concentrations. Most lead concentrations in excess of the then MAC were between 100 and 250µg/l, which is much lower than the peaks found in 1988.

A comparison of random daytime sample (slow flow) with flushed samples (fast flow) was undertaken with the findings as given in Tables B.17 and B.18.

Table B.17 - Comparison of random daytime (rdt) and flushed samples - 1989 survey

SAMPLE	LEAD % OF SAMPLES	IRON % OF SAMPLES	ALUMINIUM % OF SAMPLES
Flushed > rdt	9.2	36.0	44.5
Flushed < rdt	60.7	56.1	46.6
Flushed = rdt	30.1	7.9	8.9

Table B.18 - Comparison of random daytime (rdt) and flushed samples with lead concentrations exceeding 100µg/l - 1989 survey

SAMPLE	LEAD % OF SAMPLES	IRON % OF SAMPLES	ALUMINIUM % OF SAMPLES
Flushed > rdt	20.0	33.3	53.3
Flushed < rdt	80.0	60.0	46.7
Flushed = rdt	0.0	6.7	0.0

B.6 Comparison of 1984, 1988 and 1989 Surveys

Table B.19 shows that the overall iron concentrations in problem years are significantly higher than in non-problem years. In 1984 and 1988 exceedence of MAC for iron (200µg/l) occurred in 12.4% and in 7.4% of the samples, while no iron concentrations >200µg/l were found in 1989. Almost all high iron values (>100µg/l) were found in premises where lead concentrations were >100µg/l, Table B.20. Less than 5% of iron values >100µg/l were found in premises where lead was absent (<20µg/l).

Table B.19 - Distribution of iron concentrations - Summary

IRON CONCENTRATION (µg/l)	PERCENTAGE OF SAMPLES		
	1984 (JULY)	1988 (APRIL)	1989 (JUNE/JULY)
≤50	48.4	79.5	77.5
51 - 100	30.3	8.9	19.2
101 - 200	8.9	4.2	3.3
201 - 500	7.1	3.7	0.0
>500	5.3	3.7	0.0

Table B.20 - Number and percentage of samples with iron concentration exceeding 100µg/l in various categories of lead concentrations - Summary

LEAD CONCENTRATION (µg/l)	SAMPLES WITH IRON CONCENTRATION >100µg/l					
	1984		1988		1989	
	No.	%	No.	%	No.	%
≤20	1 of 44	2.3	8 of 161	5.0	7 of 187	3.7
21 - 50	1 of 32	3.1	2 of 11	18.2	1 of 37	2.7
51 - 100	2 of 11	18.2	0 of 6	0.0	0 of 9	0.0
101 - 200	9 of 11	81.8	1 of 1	100.0	0 of 3	0.0
>200	12 of 13	92.3	11 of 11	100.0	1 of 3	33.3

Aluminium concentrations in 1984 and 1988 were significantly higher than in 1989. In 1988 aluminium was also significantly higher than in 1984 (Table B.21).

Table B.21 - Distribution of aluminium concentrations - Summary

ALUMINIUM CONCENTRATION (µg/l)	PERCENTAGE OF SAMPLES		
	1984 (JULY)	1988 (APRIL)	1989 (JUNE/JULY)
≤50	4.5	0.0	47.0
51 - 100	85.7	74.7	43.0
>100	9.8	25.3	10.0

In 1984 only 2.3% of aluminium values >100µg/l were found in samples from premises where lead was absent (1 out of 44 premises), while more than 60% of premises with high lead concentrations (>100µg/l) also had high aluminium concentrations (see Table B.22). The distribution of high aluminium concentrations is very similar to the distribution of high iron concentrations for this year, 1984 (see Tables B.19 and B.21).

In 1988, on the other hand, high aluminium concentrations were also found in many premises where lead was absent (<20µg/l, see Table B.22), nearly a quarter of these premises had aluminium concentrations >100µg/l. The distribution of high aluminium is quite different from the distribution of high iron values in 1988. This indicates that problems with increased aluminium were more or less restricted to lead problem houses in 1984; in 1988 the increased aluminium levels were more widespread. The earlier sampling date in 1988 has to be taken into account.

Table B.22 - Number and percentage of samples with aluminium concentrations exceeding 100µg/l in various categories of lead concentrations - Summary

LEAD CONCENTRATION (µg/l)	SAMPLES WITH ALUMINIUM CONCENTRATION >100µg/l					
	1984		1988		1989	
	No.	%	No.	%	No.	%
≤20	1 of 44	2.3	36 of 161	22.4	23 of 187	12.3
21 - 50	1 of 32	3.1	1 of 11	9.1	3 of 37	8.1
51 - 100	1 of 11	9.1	3 of 6	50.0	0 of 9	0.0
101 - 200	0 of 11	0.0	1 of 1	100.0	0 of 3	0.0
>200	8 of 13	61.5	7 of 11	63.6	1 of 3	33.3

An overview of the results is presented in Table B.23.

Table B.23 - Overview of survey results

	1984	1988	1989
Sample date	July	April	June/July
Lead problem identified	Yes	Yes	No
LEAD:			
rdt/flushed where lead >100µg/l	-	flushed >rdt	flushed <rdt
High lead associated with:	High iron/aluminium sometimes manganese	High iron/aluminium sometimes manganese	Hardly any high lead
IRON:			
rdt/flushed where lead >100µg/l	-	flushed >rdt	flushed <rdt
High iron >100µg/l	Predominantly in high lead premises. Hardly any high iron where lead is low (<20µg/l)		Hardly any high iron
ALUMINIUM:			
rdt/flushed where lead >100µg/l	-	flushed >rdt	Not consistent
High aluminium >100µg/l	Predominantly in high lead premises. Hardly any high aluminium when lead is low	In premises with no lead and with high lead (more in high lead houses)	Hardly any high lead, some high aluminium in premises with no lead
RATIO LEAD:IRON	About 1.5	About 1.5 (1.1 - 2.5)	Erratic
OBSERVATIONS:	-	-	Extremely hot summer

APPENDIX C

REPORT ON INVESTIGATIONS INTO FACTORS AFFECTING PARTICULATE LEAD IN AREA C TAPWATER (PIPE RIG)

APPENDIX C

REPORT ON INVESTIGATIONS INTO FACTORS AFFECTING PARTICULATE LEAD IN AREA C TAPWATER (PIPE RIG)

S M Williams

A D Hulsmann

SUMMARY

I OBJECT

The objectives of this project were to identify the factors that affect lead concentrations in Area C tapwater and to test the effectiveness of remedial actions, in particular the effect of improved water treatment. A further objective was to assess the optimum alkalinity and pH that, together with the improved water treatment, resulted in the lowest lead levels.

II REASONS

There was a requirement by HM Government to the European Commission that water supplies will satisfy the requirements of the EC Directive relating to the quality of water for Human Consumption. (This has been superseded by the Water Regulations (1989).) In the case of soluble lead, problem areas have been identified, and successful remedies have been developed and applied. However, insoluble colloidal particles of lead corrosion products have been identified in Area C tap water, and at various other locations throughout the UK. These occur despite the application of remedies developed for soluble lead problems. It is therefore necessary to understand why particulate lead occurs and to investigate potential remedies.

III CONCLUSIONS

The overall conclusions of this work were that the joint presence of iron and organic matter could be a significant cause of elevated lead levels in the existing Area C tapwater. Improved water treatment, which removed both the iron and the organic matter, significantly decreased lead levels. Changing the alkalinity of this improved quality water (within the range 16-50mg/l as $CaCO_3$) did not have a significant effect on lead levels, neither was there conclusive evidence that silicate or phosphate dosing affected lead concentrations. Changing the pH within the range 7.5 - 9.0 did not affect lead levels.

There were occasional very high lead levels, both with and without improved water treatment. This implies that the potential for high lead levels caused by physical disturbance of the lead pipes persists, despite improved water treatment.

IV RECOMMENDATIONS

The improved water treatment to remove iron and organic matter, should result in reduced lead levels in distribution, regardless of the alkalinity or pH of the final water. It is recommended that monitoring of lead in distribution is carried out, both before and during the introduction of the improved quality treated water, to ensure that anticipated reductions in lead levels do indeed occur. If problems persist and are related to elevated iron levels, it is suggested that silicate or polyphosphate dosing is performed on a trial basis.

V RESUMÉ OF CONTENTS

This report describes the results of lead pipe rig trials that have been performed at the treatment works supplying Area C to assess factors which affect lead levels in Area C tapwater. A brief description of the experimental procedure is given. The results are discussed and conclusions drawn.

CONTENTS

1. INTRODUCTION

1.1 Objectives

The objectives of this project were to identify the factors that affect lead concentrations in Area C tapwater and to test the effectiveness of remedial actions, in particular the effect of improved water treatment. A further objective was to assess the optimum alkalinity that, together with the improved water treatment, resulted in the lowest lead levels.

1.2 Experimental procedure

In order to achieve these objectives a pipe rig consisting of eight lead pipes, each two metres long, was constructed at the treatment works supplying the area. The pipes were exhumed service pipes from Area C, four of which had been cleaned using compressed CO_2 and four which had not been cleaned. A series of experiments was performed, and the quality of the water flowing through the lead pipes was altered, as described in Section 1.3. Each experiment involved collecting water samples from each of the eight pipes for analysis for lead, iron and pH. Samples were taken at four time intervals; after flushing, and after ½, 2 and 18+ hours stagnation in the lead pipes. Thus lead concentration versus time curves (stagnation curves) were obtained for replicate pipes supplied with the various water qualities under test.

1.3 Timetable

The timetable for the experiments is listed in Table 1. Initially the rig was located within the treatment works and a period of over six months was allowed for the lead pipes to stabilise on the old water. Half-hour stagnation samples were taken regularly to assess whether the lead levels had stabilised. Once it had been established that the rig was stable the experiments with the old water were carried out. These consisted of assessing the effect of iron and Kalipol addition on lead levels in old water.

Table 1 - Timetable of experiments with the lead pipe rig

DATE		CHEMICALS ADDED	ALKALINITY (mg/l $CaCO_3$)	pH	COMMENTS
OLD WATER - HIGH IRON AND ORGANICS, LOW ALKALINITY					
Aug 88		-	<10	8.4	Intermittent water flow
Aug 88 and April 89		-	<10	8.3 - 9.5	Repeated 4 times to get baseline
Aug 88		$FeCl_3$ + NaOH to give $Fe(OH)_3$ (1mg/l as Fe)	<10	7.2	Colloidal iron (2.1.3)
April 89		$FeCl_3$ (0.5mg/l as Fe)	<10	7.9	Soluble iron (2.1.3)
April 89		$FeCl_3$ (0.8mg/l as Fe)	<10	7.2	Soluble iron (2.1.3)
April 89		$FeCl_3$ (0.5mg/l as Fe)	<10	9.6	Soluble iron (2.1.3)
May 89		polyphosphate (2mg/l as P_2O_5)	<10	9.4	Kalipol (2.1.4)
NEW WATER (LOW IN IRON AND ORGANICS) IN FOUR PIPES ('NEW' PIPES). OLD WATER IN FOUR PIPES ('OLD' PIPES)					
Sept 89	New	-	30	9.0 - 9.9	Duplicated to get baseline (2.2.2)
Sept 89	New	Reduced CO_2 and lime	21	8.6 - 9.6	(2.2.5)
Sept 89	New	$Fe(OH)_3$ (0.25mg/l as Fe)	30	8.7 - 9.5	Colloidal iron (2.2.3)
Sept 89	Old	$Fe(OH)_3$ (500mg/l as Fe)	<10	7.9 - 8.5	Colloidal iron (2.2.3)
Oct 89	Old	$Fe(OH)_3$ (1.28mg/l as Fe)	<10		Colloidal iron (2.2.3)
	New Old	}orthophosphate }(0.8mg/l as P)		8.2 - 8.7 9.4	(2.2.6)

NB - figures in brackets refers to section in text

continued over

Table 1 continued

NEW WATER (LOW IN IRON AND ORGANICS) IN FOUR PIPES ('NEW' PIPES).
OLD WATER IN FOUR PIPES ('OLD' PIPES)

DATE		CHEMICALS ADDED	ALKALINITY (mg/l $CaCO_3$)	pH	COMMENTS
Oct 89	New	Old water	<10	9.4 - 9.9	Testing whether pipes have a "memory" (2.2.4) Duplicated
	Old	Old water	<10	9.4 - 9.9	
Oct-Nov 89	New	Old water	<10	8.5 - 9.0	Duplicated (2.2.5)
	Old	Old water + CO_2 + lime	30		
Nov 89	New	New water	20	9.0	Duplicated (2.2.5)
Nov 89	Old	Old water	<10	8.5 - 9.0	Duplicated (2.2.5)
Nov 89	New	New water + CO_2 + lime	30	9.0	Duplicated (2.2.5)
	New	New water + CO_2 + lime	50	9.0	Duplicated (2.2.5)
Dec 89	New	New water + silicate (1mg/l as SiO_2)	20	9.0	Duplicated (2.2.6)
Jan 90	New	New water + poly phosphate (2mg/l as P_2O_5)	20	9.0	Duplicated (2.2.6)
Jan 90	New	New water + silicate (13mg/l as SiO_2)	20	9.0	Duplicated (2.2.6)
Mar 90	New	pH varied between 7.5 and 8.5	30	7.5, 8.0, 8.5	Duplicated

NB - figures in brackets refer to section in text

The rig was then physically removed to the pilot plant and therefore a further period of stabilisation was required. When it had been established, by sampling, that the rig was stable a series of experiments was carried out with both the old and the new water (four pipes on each water). The principle factors that were tested for their effect on lead levels were:

(a) baseline with new and old water,

(b) various alkalinity new water,

(c) increased alkalinity old water,

(d) old water in the pipes stabilised on new water (to test if the pipes have a "memory") and

(e) addition of various chemical treatments (i.e. silicates and phosphates).

The various combinations of iron, organic matter and alkalinity that were tested for their effect on lead levels are shown below.

ORGANICS	ALKALINITY	COLLOIDAL IRON	DESCRIPTION
High	Low	High	Old water
Low	High	Low	New water
Low	Low	Low	New water (reduced alkalinity)
Low	High	High	New water (added iron)
High	High	High	Old water (increased alkalinity)

The effect of pH on lead levels was also studied for both old water and new water (alkalinity 30mg/l $CaCO_3$) within the pH range 7.5 - 9.0 pH units.

The statistical analysis (using a Mann Whitney U Test at the 90% confidence level) of the results obtained from these trials is described in the following sections and a full set of all the data obtained on lead levels is presented in Annex 1.

2. RESULTS

2.1 Old water

2.1.1 Intermittent flow

Initially the rig was put on a flow régime of 12 hours stagnation and 12 hours with a 2-minute flush every hour. The solenoid valve used to establish this hydraulic régime caused waterhammer in the pipes and resulted in the disturbance of the lead corrosion layer. Size-fractionation analysis of water samples from the rig showed that lead was predominantly present in large particles (1-12 and >12µm); this confirms that physical disturbance could be one of the factors affecting particulate lead levels. Size fractionation of a small number of samples from Area C showed that most lead was present in particles with sizes 0.015 - 1µm. This lead was in the same size band as the particulate/colloidal iron in the water.

The physical disturbance of the pipes caused high and random levels of particulate lead. Most of this lead was not associated with iron (iron present in fraction <1µm).

The solenoid valve was taken out and the rig was put on a continuous flow régime (1 litre per pipe per minute).

2.1.2 Baseline measurements

Base-line measurements (Annex 1 Sections 1.1 - 1.4) were done several times at various pH values. These initial experiments sometimes included additional short stagnation times. The results are presented in Table 2.

Table 2 - Baseline measurements (treated water undosed)

LEAD CONCENTRATIONS IN µg/l					
Stagnation time	4 Aug 88	17 Aug 88	12 Apr 89	26 Apr 89	Pb range
Flushed			74	86	50 - 100
5 mins	54	58			
10 mins	72	56			
15 mins	98	82			
30 mins	161	97	127	104	100 - 150
60 mins	115	120			
2 hrs		148	209	165	150 - 200
18+ hrs		690	683	573	500 - 800
pH	8.3 - 8.5	8.3 - 8.5	7.5 - 8.8*	9.5	

All data in the table are average values of 8 lead pipes.

* Water entering rig was unstable due to interruption of lime dosing. During stagnation the pH of the water increased.

2.1.3 Addition of iron

In August 1988 an experiment was carried out to measure the effect of an increased iron concentration on lead levels in water. Lead from lead pipes is thought to adsorb onto iron particles in the water and this experiment was performed to test whether this did in fact occur. $FeCl_3$ and NaOH solutions were added to the incoming water, to form colloidal $Fe(OH)_3$. Size fractionation of the chemically formed iron hydroxide confirmed that the particle size was similar to that found naturally in Area C tapwater. During this experiment the addition of the chemicals caused the pH to drop to 7.2.

A lead stagnation time curve was made which included some additional stagnation times. The results are presented in Table 3. A full set is given in Annex 1 Sections 2.1 - 2.7. The results showed that all lead concentrations in the water spiked with iron (800 - 1,000µg/l) were higher than in undosed water.

Table 3 - Lead stagnation time curves in undosed water and water spiked with colloidal iron ($Fe(OH)_3$)

LEAD CONCENTRATIONS IN µg/l		
Stagnation time	Undosed (Fe = 130µg/l)	Dosed with Fe (colloidal Fe, Fe = 1,000 µg/l
5 mins	58	77
10 mins	56	215
15 mins	82	169
30 mins	97	187
1 hr	120	280
2 hrs	148	444
18 - 23 hrs	690	1,336
pH	8.3 - 8.5	7.2

It was not clear whether the increase in lead levels were due to the pH drop or the addition of iron, therefore an additional experiment was carried out in which pH was lowered to 7.2, without the addition of iron. This showed that increased lead levels were due to the presence of particulate/colloidal iron and not an effect of a lowered pH value.

In April 1989 further experiments were carried out to study the effect of iron addition on lead concentrations. The water at that time was unstable as a result of an interruption of lime dosing. $FeCl_3$ was added to the water; this is a soluble form of iron in contrast to the colloidal iron used in earlier experiments. The pH of the final water used in the rig was 7.2 - 7.9 and pH increased during stagnation. Results in Table 4 show that addition of $FeCl_3$ at iron concentrations of 500 and 800µg/l did not have any significant effect on lead concentrations in the water, at various stagnation times.

Table 4 - Lead stagnation time curves in undosed water and water spiked with soluble iron ($FeCl_3$) at concentrations of 500 and 800µg/l as Fe

Stagnation time (hours)	LEAD CONCENTRATIONS IN µg/l		
	Undosed (base line)	500µg/l as Fe (soluble)	800µg/l as Fe (soluble)
0	74	71	79
0.5	127	118	106
2	209	189	216
18+	683	837	787
pH	7.5 - 8.8	7.9	7.2

These experiments were repeated after the lime dosing had been re-established. In April and May a lead stagnation time curve was made using undosed water at normal pH 9.6 compared with a lead stagnation time curve in water spiked with soluble iron ($FeCl_3$). Changes in pH due to addition of chemicals were corrected. No significant difference could be found between lead concentrations in undosed water and water spiked with soluble iron (see Table 5).

Table 5 - Lead stagnation time curves in undosed water and water spiked with soluble iron ($FeCl_3$, pH adjusted) 500µg as Fe

Stagnation time (hours)	LEAD CONCENTRATIONS IN µg/l	
	Undosed	Dosed with 500µg/l as Fe (soluble Fe)
0	86	50
0.5	104	55
2	165	185
18+	573	766
pH after correction	9.6	9.6

2.1.4 Addition of phosphate

Polyphosphate (Kalipol) and orthophosphate were dosed to the old water, on two separate occasions, at a concentration of 2mg/l as P_2O_5. Statistical analysis of the results summarised in Table 6 showed that the addition of both poly- and ortho-phosphate did not have a significant effect on lead concentrations. (A full set of results is given in Annex 1 Sections 3.1 - 3.3).

Table 6 - Lead stagnation time curves for undosed water and water dosed with phosphate

Stagnation time (hours)	LEAD CONCENTRATIONS (µg/l)		
	Undosed	Dosed with Kalipol ($2mg/l\ P_2O_5$)	Dosed with orthophosphate ($2mg/l$ as P_2O_5)
0	86	70	193
0.5	104	89	185
2	165	239	240
18+	573	767	633
pH	9.6	9.4	9.4

2.2 Comparison of old and new water

2.2.1 Water quality

The main difference in quality between the old and new water is that in the new water the iron concentration, colour and organic content are reduced (compared with the old water) and the alkalinity can be increased, by CO_2 and lime dosing, to the alkalinity that produces the lowest lead levels and has the least effect on the leaching of lime from cement mortar linings. Table 7 summarises the main differences in quality produced by the different water treatments.

Table 7 - Characteristics of old and new water

DETERMINAND	OLD	NEW
pH	8.5 - 9.0	9.0
Fe (mean µg/l)	150 - 300	<50
Alkalinity (mg/l as $CaCO_3$)	<10	Can be varied between about 16-50
Organics and colour	High	Low

2.2.2 Baseline measurements

Once the rig had stabilised in its new location a duplicate set of stagnation curves was obtained, with four pipes conveying old water and four conveying new water. The results are summarised in Table 8 (and Annex 1 Sections 4.1 - 4.4) and show that the new water produces much lower lead levels than the old water. The decrease in lead concentration is most obvious at longer stagnation times. Another interesting feature of this set of results is that the lead levels in the flushed samples were on average higher than those in the half-hour stagnation samples. This result indicates that the mechanism for the occurrence of high lead levels is likely to be a combination of a scouring and an adsorption effect.

Table 8 - Lead concentrations and percentage decrease between old and new waters

Stagnation time (hours)	AVERAGE LEAD CONCENTRATIONS (µg/l)		% decrease in lead
	Old	New	
0	127	70	45%
0.5	124	47	62%
2	700	152	78%
18+	925	167	82%
Average iron (µg/l)	462	19	
Alkalinity (mg/l $CaCO_3$)	<10	30	

2.2.3 Addition of iron

The decrease in lead in the final water with improved treatment could be caused by higher alkalinity, lower iron content, lower organic content or lower colour of the new water. To identify the factor that caused the decrease in lead concentrations some experiments were carried out in which the colloidal iron concentration in the old and new water was increased.

The results of the experiments with the addition of colloidal iron are summarised in Table 9 (Annex 1 Sections 4.1 - 4.4 and 5.1 - 5.3).

Table 9 - Comparison of lead levels for old and new water with and without the addition of colloidal iron

Stagnation time (hours)	AVERAGE LEAD CONCENTRATIONS (µg/l)						
	OLD WATER				NEW WATER		
	Undosed		Dosed with $Fe(OH)_3$		Undosed		Dosed with $Fe(OH)_3$
0	165	90	157	130	75	85	62
0.5	147	100	562	135	55	60	57
2	-	700	1625	497	-	157	117
18+	1160	690	6325	-	210	125	212
Average iron (mg/l)	0.52	0.41	517*	1.28	0.025	0.015	0.25

* An unintentional overdose of $Fe(OH)_3$ entered the four pipes supplied with old water. However the results proved interesting and have therefore been included.

The comparison between lead concentrations in old water undosed and old water dosed with particulate iron is difficult because of incomplete data (as no long stagnation time was carried out in one experiment, there are not sufficient data for a statistical analysis). Comparison of lead concentrations at short stagnation times does not show a significant difference between dosed and undosed old water. This is contrary to the findings outlined in Section 2.1.3. During one of the experiments the incoming water was overdosed by mistake with particulate iron, resulting in an average iron concentration of 517mg/l. It is interesting to note the dramatic effect on lead concentrations especially at the longer stagnation times. Even though these iron concentrations are unrealistically high, it indicates the effect disturbances in a cast-iron system might have on lead concentrations.

Statistical analysis of the new water spiked with 0.25mg/l of iron showed that the iron had no significant effect on lead concentrations.

2.2.4 Memory effect

During the course of these lead pipe rig experiments there was concern that, as the quality of the water being supplied to the lead pipes was being changed at frequent intervals, this may influence the results of the trials. A set of duplicate experiments was therefore performed to establish whether the pipes in the rig had a 'memory'; (i.e. did the water quality from the previous experiment affect the results of the current experiment).

In order to test whether the lead pipes had a memory, 'old' water was supplied to the pipes that had been receiving 'new' water. The 'old' pipes continued to receive 'old' water and served as controls.

The results are summarised in Table 10 (Annex 1 Sections 6.1 - 6.4).

Table 10 - Comparison of lead levels between pipes supplied with old water with half conditioned on new water and half conditioned on old water

Stagnation Time (hours)	AVERAGE LEAD CONCENTRATIONS (µg/l)			
	Old water in old pipes		Old water in new pipes	
	I	II	I	II
0	1778	418	4308	750
0.5	383	89	1265	345
2	233	130	233	198
18+	1035	343	1118	435
Average iron (mg/l)	0.87	0.33	0.57	0.34

The first experiment showed no significant difference in lead concentrations between pipes conditioned on old and those conditioned on new water (Mann Whitney U Test, 90% confidence interval). In the second experiment the 'new' pipes showed significantly higher lead concentrations than the 'old' pipes, when both were fed with old water. Where only the long stagnation times were considered there was no significant difference between 'old' and 'new' pipes. From this and the fact that the first experiment did not show a difference between 'old' and 'new' pipes (lead concentrations increased immediately), it was concluded that there is no memory effect. Hence it was concluded that the pipes did not have a memory and it was therefore valid to perform a sequence of experiments without allowing a period in between experiments.

2.2.5 Variation in alkalinity

The baseline experiments described in Section 2.2.2 showed that old water passing through lead pipes produced higher lead levels than new water. The cause could either have been the difference in alkalinity, iron or organic content. In order to test whether it was the alkalinity, old water alkalinity was increased with CO_2 and lime to 30mg/l (as $CaCO_3$). Half of the pipes continued to receive old water with normal alkalinity (<10mg/l) and these were used as controls. The results are summarised in Table 11 (Annex 1 Sections 7.1 - 7.4).

Table 11 - Comparison of lead levels between old water and old water with increased alkalinity

Stagnation Time (hours)	AVERAGE LEAD CONCENTRATIONS (µg/l)			
	Old water (alkalinity <10mg as $CaCO_3$)		Old water + CO_2 and lime (alkalinity 30mg/l as $CaCO_3$)	
	I	II	I	II
0	823	58	158	163
0.5	225	60	205	85
2	130	93	158	545
18+	328	215	473	498

In the first experiment there was no significant difference in lead concentration as a result of increased alkalinity. In the second experiment the old water with increased alkalinity showed significantly higher lead concentrations than the control. However, comparison with other control data (experiments with old water in old pipes) showed that lead concentrations in this particular experiment were extremely low (significantly lower than duplicate results). A statistical analysis was therefore carried out using all the baseline data for old water in old pipes and comparing this with the data for old water with increased alkalinity. This analysis showed increasing the alkalinity of the old water does not have a significant effect on lead levels when compared with lead levels in low alkalinity old water. Hence it can be concluded that it was not the increased alkalinity that caused the lower lead concentrations in the new water.

In order to test whether there was an optimum alkalinity, of the new water, which produced the lowest lead levels, a series of experiments was carried out. The alkalinity of the new water was varied between 16 and 50mg/l (as $CaCO_3$). The results of the lead concentrations in the various different alkalinity waters were compared with each other and with lead levels from the old water. The results are summarised in Table 12 (Annex 1 Sections 8.1 - 8.14).

Table 12 - Comparisons of lead levels between old water and various alkalinity new waters

	AVERAGE LEAD LEVELS (µg/l)					
Stagnation Time (hours)	Alkalinity (mg/l as $CaCO_3$)					
	New water					Old water
	16[*]	21[*]	20[¶]	30[$]	50[¶]	<10[α]
0	50	55	72	77	107	364
0.5	50	52	45	77	74	444
2	135	77	74	102	74	325
18+	-	155	121	124	120	587

[*] average of 1 stagnation curve for 4 pipes
[¶] average of 2 stagnation curves for 4 pipes
[$] average of 4 stagnation curves for 4 pipes
[α] average of 9 stagnation curves for 4 pipes

Statistical analysis of the complete set of results showed that there was no significant difference in lead levels between the new water at the various alkalinities, within the range 16-50mg/l as $CaCO_3$. All experiments with the new water showed significantly lower lead concentrations than those measured in the old water.

2.2.6 Addition of phosphate and silicate

Orthophosphate and polyphosphate were both dosed to the new water at a dose rate of 2mg/l (as P_2O_5). Sodium silicate was dosed at two concentrations, 1 and 13mg/l (as SiO_2). The results are summarised in Table 13 (and shown in full in Annex 1 Sections 9 and 10).

Table 13 - Average lead levels in new water dosed with silicate or phosphate

Stagnation Time (hours)	AVERAGE LEAD CONCENTRATIONS (µg/l)						
	SiO_2 (1mg/l)	SiO_2 (1mg/l)	SiO_2 (13mg/l)	SiO_2 (13mg/l)	Ortho-phosphate (2mg/l)	Poly-phosphate (2mg/l)	Poly-phosphate (2mg/l)
0	37	91	60	108	118	95	60
½	46	50	103	<50	83	128	50
2	80	31	63	55	65	108	63
18+	65	65	58	55	78	123	60

These results were compared with various experiments on the new water, without any chemical dosing. The results of the "control" experiments are listed in Table 14.

Table 14 - Average lead concentrations in new water without chemical dosing

Stagnation Time (hours)	LEAD CONCENTRATIONS µg/l							
	Alkalinity of new water (mg/l $CaCO_3$)							
	20	20	30	30	30	21	50	50
0	85	60	128	21	55	85	175	39
½	53	38	160	35	52	60	105	44
2	83	66	88	60	77	158	80	69
18+	188	55	85	82	155	125	158	83

Comparisons were made between the results of lead concentrations with and without silicate dosing. A total of 32 comparisons were possible. The results showed that in thirteen cases, dosing resulted in significantly lower lead concentrations than the control experiments. However in nineteen cases, there was no significant difference in lead concentrations between dosed and undosed waters. However, three of the control experiments were significantly higher than the remaining control experiments. It was the comparison of the results from these three experiments with the lead levels during silicate dosing that resulted in apparently lower lead concentrations with sodium silicate. Hence, because of the variability in the control data there is no conclusive evidence that sodium silicate reduces lead levels. What is clear from the results is that there is no significant difference between lead levels in the waters dosed with high and low silicate dose. As the great difference in silicate dose does not affect lead concentrations this would seem to add weight to the argument that dosing silicate does not have a significant effect on lead concentrations.

The comparison between lead concentrations with and without orthophosphate dosing revealed that in six cases there was no significant difference, but in two cases (where control lead concentrations were high), the orthophosphate dosing appeared to cause a significant reduction in lead levels. Again, the conclusion has to be that the rig results show that orthophosphate has no significant effect on lead levels.

The result for polyphosphate was similar to that for orthophosphate with ten comparisons showing no significant difference between lead levels in dosed and undosed water, and three each showing a significant decrease and a significant increase respectively. Thus the conclusion is as with both silicate and orthophosphate, that there is no conclusive evidence to show that these treatment chemicals affect lead levels.

3. DISCUSSION

The objectives of this project were to identify the factors that affect lead concentrations in Area C tapwater and to investigate the effects of remedial actions.

In order to achieve these objectives a lead pipe rig was constructed and a series of experiments was performed with various quality waters. During the course of these experiments a set of trials was carried out to assess whether the lead pipes had a "memory" (i.e. did the water quality from the previous experiment affect the results of the current experiment). The results from this test showed that the lead pipes had no memory and it was thus concluded that it was valid to perform a sequence of experiments without allowing a period for stabilisation in between experiments.

It is clear, from the results presented, that treating "old" water to reduce iron and organic matter and slightly increase the alkalinity, results in consistently low lead levels.

Several experiments have been performed to assess the factors which affect lead concentrations in Area C water. At the early stages of the experimental work it became clear that physical disturbance of the lead pipework caused variable and high lead levels. Under these circumstances it was ascertained that the lead was present in large particles, which were not associated with iron. Following these results, attempts were made to keep physical disturbance of the lead pipes to a minimum.

The addition of soluble iron to old water had no significant effect on lead levels. One experiment in which insoluble iron (iron hydroxide) was dosed to old water showed that this caused a significant increase in lead levels. However this was not reproduced when the experiment was repeated. Furthermore the addition of insoluble iron to new water also had no significant effect on lead concentrations. These results suggest that in some cases the presence of iron can cause increased lead concentrations but it is likely that other factors, such as the presence of organic matter, are involved as well. This would explain the inability to duplicate the experimental results by spiking the water with chemically produced iron.

Further evidence that the combination of iron and organic matter caused the elevated lead levels was obtained when the alkalinity of the old water was increased to the level of that in the new water. At this stage the difference in quality between the two waters was the levels of iron and organic matter. Nevertheless the lead levels in new water were significantly lower than in old water with increased alkalinity. Also there was no significant difference between lead levels in old water and those in old water with increased alkalinity. Hence it can be concluded that the decrease in lead levels in new water must be caused by the absence of iron and organic matter. A summary of the various experiments in which iron, alkalinity and organic matter were varied is shown below in Table 15.

Table 15 - Summary of the effect on lead levels of varying the organics, iron and alkalinity of Area C tapwater

DESCRIPTION	ORGANICS	ALKALINITY	COLLOIDAL IRON	RESULTANT LEAD LEVELS
Old water	H	L	H	H
New water	L	H	L	L
New water, reduced alkalinity	L	L	L	L
New water, added iron	L	H	H	L
Old water, increased alkalinity	H	H	H	H

H = High, L = Low

Varying the alkalinity of new water within the range 16-50mg/l (as $CaCO_3$) did not show a significant effect on lead levels. Although the lead levels in all cases were significantly lower than those measured in the old water, they were unaffected by changes in alkalinity. Thus it can be concluded that, for the purposes of lowering lead levels, the alkalinity of the new water is unimportant (within the range 16-50mg/l as $CaCO_3$).

Dosing of silicate, orthophosphate and polyphosphate to the new water did not appear to cause a reduction in lead levels on the rig. However this should not rule out the possibility of their application into the distribution system. It is possible that, when new water is introduced into distribution, although iron concentrations in the final water may be low, there may be residual iron and organic matter in the deposits in the mains which may persist in causing some instances of elevated lead at the tap. However as a high percentage of the mains in Area C have been relined this is unlikely to be the case.

If this does occur it is possible that chemical dosing may work indirectly to lower lead levels by lowering the particulate/colloidal iron levels in the water entering the service pipe. The use of these chemicals should therefore still be considered if elevated lead levels persist following the introduction of new water into distribution.

4. CONCLUSIONS

The overall conclusions of this project are that the factors that affect particulate lead concentrations in Area C tapwater are (a) the joint presence of particulate/colloidal iron and organic matter and (b) physical disturbance of the internal corrosion layer. Treating water to remove the iron and organic matter results in a significant reduction in lead levels. This reduction in lead levels is unaffected by variations in alkalinity.

In reaching these overall conclusions, the following conclusions have also been drawn.

1. Changing the alkalinity of the new water (within the range 16-50mg/l $CaCO_3$) has no significant effect on lead levels. Neither does the addition of particulate iron or changing the pH within the range 7.5 - 9.0.

2. In old water varying the following parameters has no significant effect on lead levels.

   alkalinity (30mg/l as $CaCO_3$)
   addition of soluble iron (0.5-0.8mg/l as Fe)
   pH (7.6 - 9.6).

3. Addition of particulate iron to old water sometimes results in significantly higher lead levels compared with those measured in old water without added iron.

4. A potential contributor to high lead levels in tapwater is physical disturbance of the lead pipes. When this occurs the lead is present in large particles and is not associated with iron.

5. The lead pipes in the pipe rig respond quickly to a change in water quality. It is thus valid to perform a series of experiments, with different quality waters, without allowing for a period of stabilisation between experiments.

6. Results from the pipe rig experiments suggest that chemical dosing (silicate and phosphate) has no significant effect on lead concentrations.

5. RECOMMENDATIONS

The improved water treatment to remove iron and organic matter should result in reduced lead levels in distribution, regardless of the alkalinity or pH of the final water (within the ranges tested). It is recommended that monitoring of lead in distribution is carried out, both before and during the introduction of the improved quality treated water, to ensure that anticipated reductions in lead levels do indeed occur. If problems persist and are related to elevated iron levels, it is suggested that silicate or polyphosphate dosing is performed on a trial basis.

ANNEX 1

LEAD RESULTS FROM STAGNATION CURVES

1. BASELINE MEASUREMENTS OLD WATER

1.1

4 AUG 1988	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0.08	<50	<50	60	<50	63	<50	60	<50	54
0.16	54	69	86	<50	80	66	<50	116	72
0.25	68	76	86	53	83	<50	290	76	98
0.5	60	104	85	101	<50	123	678	89	161
1	24500*	130	147	75	<50	83	220	99	115
pH range 8.3 - 8.5									

1.2

17 AUG 1988	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0.08	<50	<50	116	<50	<50	<50	450	<50	58
0.16	<50	69	<50	66	<50	<50	<50	59	56
0.25	114	<50	<50	<50	<50	<50	<50	242	82
0.5	60	136	89	<50	<50	78	60	254	97
1	90	114	136	80	<50	113	80	294	120
2	121	271	260	<50	53	84	144	200	148
18+	518	371	1620	264	826	492	739	4590*	690
pH range 8.3 - 8.5									

*** Outliers - excluded from statistical analysis**

1.3

12 APR 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO STAGNATION TIME (HRS)	1	2	3	4	5	6	7	8	Mean
0	95	106	338*	94	57	55	60	<50	74
0.5	109	82	219	79	72	138	227	91	127
2	-	227	295	189	109	250	214	179	209
18+	272	961	1320	255	181	1050	949	473	683
pH range 7.5 - 8.8									

1.4

26 APR 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO STAGNATION TIME (HRS)	1	2	3	4	5	6	7	8	Mean
0	<50	379*	71	175	75	90	<50	101	86
0.5	<50	190	174	108	51	101	82	78	104
2	79	342	231	145	82	177	123	141	165
18+	315	1270	717	611	156	598	552	362	573

2. OLD WATER WITH IRON ADDITION

2.1 Old water + 1mg/l colloidal iron ($Fe(OH)_3$)

17 AUG 1988	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0.08	66	99	89	101	<50	103	51	90	77
0.16	75	187	90	83	<50	1050	95	95	215
0.25	648	176	87	99	<50	99	92	106	169
0.5	213	323	140	180	56	189	145	247	187
1	248	294	421	269	357	116	238	294	280
2	379	1180	404	304	179	270	393	440	444
18+	1210	1500	1790	1450	961	1090	1190	1500	1336
pH 7.2									

2.2 Old water, undosed

17 AUG 1988	LEAD CONCENTRATION (µg/l)								
PIPE NO STAGNATION TIME (HRS)	1	2	3	4	5	6	7	8	Mean
0.08	<50	<50	116	<50	<50	<50	450	<50	58
0.16	<50	69	<50	66	<50	<50	<50	59	56
0.25	114	<50	<50	<50	<50	<50	<50	242	82
0.5	60	136	89	<50	<50	78	60	254	97
1	90	114	136	80	<50	113	80	294	120
2	121	271	260	<50	53	84	144	200	148
18+	518	371	1620	264	826	492	739	4590*	690
pH range 8.3 - 8.5									

2.3 Old water 0.5mg/l soluble iron ($FeCl_3$)

19 APR 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO STAGNATION TIME (HRS)	1	2	3	4	5	6	7	8	Mean
0	<50	60	572*	103	60	73	104	<50	71
0.5	88	126	195	127	62	164	114	72	118
2	161	187	315	179	110	225	180	156	189
18+	527	1670	1010	998	264	1150	488	589	837
pH 7.9									

2.4 Old water 0.8mg/l soluble iron ($FeCl_3$)

12 APR 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0	<50	137	77	71	<50	91	97	62	79
0.5	89	88	134	110	90	90	145	99	106
2	104	267	414	217	150	229	167	180	216
18+	461	1500	1220	896	263	1060	356	543	787
pH 7.2									

2.5 Old water, low pH

13 APR 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0	95	106	338*	94	57	55	60	<50	74
0.5	109	82	219	79	72	138	227	91	127
2	-	227	295	189	109	250	214	179	209
18+	272	961	1320	255	181	1050	949	473	683
pH range 7.5 - 8.8									

2.6 Old water pH adjusted + 0.5mg/l soluble iron ($FeCl_3$)

27 APR 89	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0	<50	<50	<50	<50	<50	<50	<50	<50	50
0.5	<50	56	77	<50	<50	<50	<50	51	55
2	139	223	277	170	109	207	182	173	185
18+	472	1340	1120	565	317	880	719	716	766
pH 9.6									

2.7 Old water undosed

26 APR 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0	<50	379*	71	175	75	90	<50	101	86
0.5	<50	190	174	108	51	101	82	78	104
2	79	342	231	145	82	177	123	141	165
18+	315	1270	717	611	156	598	552	362	573

3. OLD WATER DOSED WITH PHOSPHATE

3.1 Old water dosed with kalipol (2mg/l P_2O_5)

3 MAY 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO STAGNATION TIME (HRS)	1	2	3	4	5	6	7	8	Mean
0	59	103	89	65	<50	66	73	57	70
0.5	98	84	120	59	<50	88	98	106	89
2	219	314	298	172	124	297	233	257	239
18+	541	1550	1110	585	152	954	583	664	767
pH 9.4									

3.2 Old water dosed with orthophosphate (2mg/l P_2O_5)

17 OCT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO STAGNATION TIME (HRS)	5	6	7	8	Mean
0	100	300	80	290	193
0.5	80	180	60	420	185
2	160	480	80	220	240
18+	990	640	320	580	633
pH 9.4					

3.3 Old water undosed

26 APR 1989	LEAD CONCENTRATION (µg/l)								
PIPE NO	1	2	3	4	5	6	7	8	Mean
STAGNATION TIME (HRS)									
0	<50	379*	71	175	75	90	<50	101	86
0.5	<50	190	174	108	51	101	82	78	104
2	79	342	231	145	82	177	123	141	165
18+	315	1270	717	611	156	598	552	362	573

4. BASE-LINE OLD WATER COMPARED WITH NEW WATER

4.1 New water (alkalinity 30mg/l $CaCO_3$)

6 SEPT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	130	70	<50	<50	75
0.5	60	60	<50	<50	55
2	-	-	-	-	-
18+	380	180	180	150	210

4.2 Old water (average Fe = 520µg/l)

6 SEPT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	130	80	220	230	165
0.5	140	110	170	170	147
2	-	-	-	-	-
18+	1420	580	1240	1400	1160

4.3 New water (alkalinity 30mg/l $CaCO_3$)

11 SEPT 89	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	110	130	<50	<50	85
0.5	80	60	<50	<50	60
2	360	120	<50	100	158
18+	250	100	60	90	125

4.4 Old water (average Fe = 406µg/l)

11 SEPT 89	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	60	60	110	130	90
0.5	120	60	120	100	100
2	330	170	1380	920	700
18+	750	370	920	720	690

5. ADDITION OF IRON TO OLD AND NEW WATER

5.1 New water plus 0.25mg/l (as Fe) iron hydroxide

12 SEPT 89	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	<50	100	<50	<50	63
0.5	<50	70	<50	60	58
2	150	110	110	110	118
18+	400	180	140	140	213

5.2 Old water plus 517mg/l¶ (as Fe) iron hydroxide

14 SEPT 89	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	120	150	200	160	157
0.5	600	300	600	750	562
2	1200	710	2500	2200	1652
18+	5700	3600	8700	7300	6325

¶ Very high iron dose

5.3 Old water plus 1.28mg/l (as Fe) iron hydroxide

11 OCT 89	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	230	70	90	130	130
0.5	140	140	100	160	135
2	250	330	340	1070	497
18+	-	-	-	-	-

6. MEMORY EFFECT

6.1 Old water in "old" pipes undosed alkalinity <10mg/l as $CaCO_3$

30 OCT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO STAGNATION TIME (HRS)	5	6	7	8	Mean
0	1380	450	470	4810	1778
0.5	310	210	360	650	383
2	110	150	340	330	233
18+	360	550	600	2630	1035

6.2 Old water in 'new' pipes undosed alkalinity <10mg/l as $CaCO_3$

30 OCT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO STAGNATION TIME (HRS)	1	2	3	4	Mean
0	540	11100	1460	4130	4308
0.5	190	2400	290	2180	1265
2	280	350	100	200	233
18+	1150	1550	400	1370	1118

6.3 Old water in 'old' pipes undosed alkalinity <10mg/l as $CaCO_3$

31 OCT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	160	160	440	910	418
0.5	63	52	120	120	89
2	120	120	100	180	130
18+	510	240	260	360	343

6.4 Old water in 'new' pipes undosed alkalinity <10mg/l as $CaCO_3$

31 OCT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	1320	2260	470	150	750
0.5	360	330	100	590	345
2	190	240	110	250	198
18+	360	600	280	500	435

7. INCREASED ALKALINITY OF OLD WATER

7.1 Old water dosed with CO_2 and lime. Alkalinity 30mg/l as $CaCO_3$

02 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	690	770	1170	660	823
0.5	240	170	160	330	225
2	80	130	130	180	130
18+	280	390	300	340	328

7.2 Old water alkalinity <10mg/l as $CaCO_3$

02 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	<50	280	230	70	158
0.5	90	490	90	150	205
2	120	210	90	210	158
18+	430	640	270	550	473

7.3 Old water dosed with CO_2 and lime. Alkalinity 30mg/l as $CaCO_3$

6 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	<50	60	<50	70	58
0.5	<50	<500	60	90	63
2	100	90	100	80	93
18+	210	200	220	230	215

7.4 Old water alkalinity <10mg/l as $CaCO_3$

6 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	80	60	<50	460	163
0.5	80	90	<50	120	85
2	810	850	111	410	545
18+	430	660	320	580	498

8. VARYING ALKALINITY OF THE NEW WATER

8.1 New water alkalinity 16mg/l as $CaCO_3$

2 OCT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	<50	<50	<50	<50	50
0.5	<50	<50	<50	<50	50
2	120	140	130	150	135
18+	-	-	-	-	-

8.2 New water alkalinity 21mg/l as $CaCO_3$

14 SEP 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	<50	70	<50	<50	55
0.5	60	<50	<50	<50	52
2	130	60	<50	70	77
18+	420	70	<50	80	155

8.3 New water alkalinity 20mg/l as $CaCO_3$

9 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	<50	100	90	100	85
0.5	<50	<50	60	<50	53
2	60	70	110	90	83
18+	200	180	190	180	188

8.4 Old water alkalinity <10mg/l as $CaCO_3$

9 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	860	750	160	980	689
0.5	470	820	110	1050	613
2	160	260	60	260	185
18+	660	650	290	650	563

8.5 New water alkalinity 20mg/l as $CaCO_3$

10 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	90	49	60	40	60
0.5	29	20	37	64	38
2	70	38	47	110	66
18+	70	53	60	37	55

8.6 Old water alkalinity <10mg/l as $CaCO_3$

10 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	650	580	120	110	365
0.5	280	290	70	90	183
2	240	240	120	220	205
18+	400	420	210	420	363

8.7 New water alkalinity 30mg/l as $CaCO_3$

22 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	120	70	100	220	128
0.5	130	130	190	190	160
2	80	60	110	100	88
18+	60	<50	70	160	85

8.8 Old water alkalinity <10mg/l as $CaCO_3$

22 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	1240	1780	260	1140	1105
0.5	2010	2950	200	2460	1905
2	320	370	150	380	305
18+	340	520	230	340	358

8.9 New water alkalinity 30mg/l as $CaCO_3$

23 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	13	12	22	35	21
0.5	27	24	40	47	35
2	52	58	70	60	60
18+	71	58	60	140	82

8.10 Old water alkalinity <10mg/l as $CaCO_3$

23 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	90	190	45	110	109
0.5	80	90	88	120	95
2	150	190	120	240	175
18+	270	390	170	380	303

8.11 New water alkalinity 50mg/l as $CaCO_3$

16 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	230	160	130	180	175
0.5	230	51	90	48	105
2	60	60	70	130	80
18+	210	120	130	170	158

8.12 Old water alkalinity <20mg/l as $CaCO_3$

16 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	310	1100	120	238	442
0.5	610	2060	130	100	725
2	200	371	80	210	215
18+	660	1820	440	890	953

8.13 New water alkalinity 50mg/l as $CaCO_3$

17 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	35	34	53	33	39
0.5	39	30	59	47	44
2	65	60	80	70	69
18+	90	70	80	90	83

8.14 Old water, undosed, alkalinity <20mg/l as $CaCO_3$

17 NOV 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	440	130	60	-	210
0.5	120	100	<50	140	103
2	130	430	110	220	223
18+	280	-	190	390	287

9. SILICATE DOSING NEW WATER

9.1 New water dosed with 1mg/l (as SiO_2) sodium silicate

14 DEC 1989	LEAD CONCENTRATION (μg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	6	<50	<50	40	37
0.5	23	<50	<50	62	46
2	36	110	100	70	80
18+	60	70	70	56	65

9.2 Old water undosed

14 DEC 1989	LEAD CONCENTRATION (μg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	450	1010	80	160	425
0.5	130	190	90	100	128
2	240	190	100	160	173
18+	320	-	210	300	277

9.3 New water dosed with 1mg/l (as SiO_2) sodium silicate

20 DEC 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	130	110	22	100	91
0.5	90	17	36	58	50
2	31	25	33	33	31
18+	70	70	<50	70	65

9.4 Old water undosed

20 DEC 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	400	490	80	520	373
0.5	240	220	57	40	140
2	130	150	120	-	133
18+	360	390	260	460	368

9.5 New water dosed with 13mg/l (as SiO_2) sodium silicate

24 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	<50	<50	90	<50	60
0.5	160	<50	150	<50	103
2	60	<50	80	60	63
18+	<50	<50	70	60	58

9.6 Old water undosed

24 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	100	190	<50	740	270
0.5	850	1900	160	1370	1070
2	240	450	120	240	263
18+	250	510	110	500	343

9.7 New water dosed with 13mg/l (as SiO_2) sodium silicate

25 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	120	120	130	60	108
0.5	<50	<50	<50	<50	50
2	70	<50	<50	<50	55
18+	<50	60	60	<50	55

9.8 Old water undosed

25 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	450	670	110	280	378
0.5	70	80	<50	70	68
2	140	210	140	180	168
18+	240	420	190	270	280

10. PHOSPHATE DOSING NEW WATER

10.1 New water alkalinity 30mg/l as $CaCO_3$ dosed with kalipol 2mg/l (as P_2O_5)

10 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	<50	<50	<50	230	95
0.5	70	<50	200	190	128
2	90	70	100	170	108
18+	60	250	110	70	123

10.2 Old water alkalinity <10mg/l, as $CaCO_3$ undosed

10 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	80	340	<50	390	215
0.5	770	1470	120	1150	878
2	280	330	130	400	265
18+	970	320	80	140	428

10.3 New water alkalinity 30mg/l as $CaCO_3$ dosed with kalipol 2mg/l (as P_2O_5)

11 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	5	6	7	8	Mean
STAGNATION TIME (HRS)					
0	80	60	<50	<50	60
0.5	<50	<50	<50	<50	50
2	<50	<50	90	60	63
18+	<50	<50	60	80	60

10.4 Old water alkalinity <10mg/l, as $CaCO_3$ undosed

11 JAN 1990	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	750	90	<50	<50	270
0.5	80	170	60	80	98
2	120	190	90	120	130
18+	270	310	160	250	247

10.5 New water dosed with orthphosphate (2mg/l as P_2O_5)

17 OCT 1989	LEAD CONCENTRATION (µg/l)				
PIPE NO	1	2	3	4	Mean
STAGNATION TIME (HRS)					
0	<50	<50	230	140	118
0.5	<50	<50	180	<50	83
2	<50	<50	110	<50	65
18+	70	<50	90	100	78

APPENDIX D

SUMMARY OF TRIALS WITH REMEDIAL ACTIONS INTENDED TO REDUCE LEAD LEVELS IN PART OF THE NORTH OF ENGLAND

APPENDIX D

SUMMARY OF TRIALS WITH REMEDIAL ACTIONS INTENDED TO REDUCE LEAD LEVELS IN PART OF THE NORTH OF ENGLAND

S M Williams

1. - AREA E

1.1 Summary

Area E is an area suffering from particulate lead problems. Orthophosphate has been dosed at a rate of 1.2mg/l as P since March 1989. This has reduced average lead levels but has not reduced peaks in lead concentrations.

1.2 Results

Table 1 - Distribution of lead and iron concentrations during surveys in Area E

Lead					
Concentration range (µg/l)	Nov 87 No dosing	May 88 No dosing	Feb 89 No dosing	Apr 89 Orthophosphate	July 89 Orthophosphate
0 - 25	70%	70%	87%	84%	78%
26 - 50	20%	16%	8%	3%	11%
51 - 75	7%	5%	4%	5%	4%
76 - 100	1%	4%	0%	2%	4%
> 100	2%	5%	1%	6%	3%
Average lead* (µg/l)	21	19	13	13	17
Iron					
Concentration range (µg/l)	Nov 87 No dosing	May 88 No dosing	Feb 89 No dosing	Apr 89 Orthophosphate	July 89 Orthophosphate
0 - 25	-	3%	0%	3%	28%
26 - 50	-	40%	25%	48%	53%
51 - 75	-	37%	46%	33%	12%
76 - 100	-	9%	15%	4%	4%
> 100	-	11%	14%	12%	3%
Average iron (µg/l)	-	66	69	58	40

* average lead concentrations excluding lead concentrations ≥100µg/l

The measured changes in lead and iron levels have been tested for significance using a Mann-Whitney U test at the 95% confidence level. The actual changes have been compared with the anticipated seasonal changes. This is shown in Table 2.

Table 2 - Results of statistical analysis of the effect of orthophosphate dosing on lead and iron concentrations

DATES	EXPECTED CHANGE LEAD AND IRON	MEASURED CHANGE LEAD	MEASURED CHANGE IRON
Nov 87 - Apr 89	no change	decrease	no data
Nov 87 - Jul 89	increase	decrease	no data
May 88 - Apr 89	no change	decrease	no change
May 88 - Jul 89	increase	no change	decrease
Feb 89 - Apr 89	increase	no change	decrease
Feb 89 - Jul 89	increase	no change	decrease

1.3 Conclusions

Orthophosphate dosing has significantly reduced average lead concentrations, particularly when compared with the anticipated seasonal changes. However dosing has been unsuccessful at achieving compliance with the requirements of EAGIM surveys. Nevertheless, the expected seasonal increase in peaks of lead concentrations does not occur. Orthophosphate dosing has resulted in reduced average iron concentrations and reduced peaks in iron concentrations.

2. AREA D

2.1 Summary

Area D has been identified as a zone with a combination of a particulate and a soluble lead problem. Trials with Kalipol and orthophosphate have been carried out. Kalipol did not reduce lead levels and orthophosphate did.

2.2 Results

Table 3 - Distribution of lead and iron concentrations during surveys in Area D

Lead					
Concentration range (µg/l)	Feb 88 No dosing	July 88 Kalipol	Dec 88 Orthophosphate	Jan 89 Orthophosphate	May 89 Orthophosphate
0 - 25	70%	63%	89%	94%	89%
26 - 50	18%	18%	10%	3%	5%
51 - 75	4%	7%	0%	0%	4%
76 - 100	1%	6%	1%	2%	1%
> 100	5%	5%	0%	1%	1%
Average lead* (µg/l)	18	25	11	13	15

Iron					
Concentration range (µg/l)	Feb 88 No dosing	July 88 Kalipol	Dec 88 Orthophosphate	Jan 89 Orthophosphate	May 89 Orthophosphate
0 - 25	-	-	12%	20%	10%
26 - 50	-	-	63%	53%	54%
51 - 75	-	-	19%	17%	23%
76 - 100	-	-	4%	6%	8%
> 100	-	-	2%	4%	5%
Average iron (µg/l)	-	-	45	43	49

* average lead concentrations excluding lead concentrations ≥100µg/l

Table 4 - Results of statistical analysis of the effect of poly- and orthophosphate dosing on lead levels

DATES		EXPECTED CHANGE	MEASURED CHANGE
No Dose -	Poly P		
Feb 88	July 88	increase	increase
No Dose -	Ortho P		
Feb 88	Nov/Dec 88	no change	decrease
Feb 88	Jan 89	no change	decrease
Feb 88	May 89	increase	decrease
Poly P -	Ortho P		
July 88	Nov/Dec 88	decrease	decrease
July 88	Jan 89	decrease	decrease
July 88	May 89	decrease	decrease

2.3 Conclusions

Dosing polyphosphate showed no beneficial effect on lead concentrations (from limited data). No iron data are available to assess the effect of polyphosphate on iron. Dosing orthophosphate gave a significant decrease in lead concentrations. The effect on iron is unknown as no iron data are available for period before dosing.

3. AREA F

3.1 Summary

There is a particulate lead problem in Area F. Polyphosphate dosing was applied in May 1988 at a dose of 1 mg/l as P. This reduced iron levels and also reduced average lead levels compared with the anticipated seasonal trends in lead concentrations. However, the number of peaks in lead levels was not reduced by Kalipol dosing and therefore the treatment failed to bring the area into compliance with the requirements of the EAGIM surveys. Orthophosphate was therefore dosed, commencing in April 1989 at a dose rate of 1 mg/l as P. It is too early to tell whether this has been effective.

Table 5 - Distribution of lead and iron concentrations during surveys in Area Fa

LEAD							
CONCENTRATION RANGE (µg/l)	OCT/NOV 87 NO DOSING	FEB 88 NO DOSING	MAY/JUNE 88 POLYPHOSPHATE	JUL 88 POLYPHOSPHATE	OCT 88 POLYPHOSPHATE	FEB 89 POLYPHOSPHATE	MAY/JUN 89 POLYPHOSPHATE
0 - 25	52%	65%	66%	60%	76%	68%	64%
26 - 50	28%	23%	19%	23%	15%	18%	22%
51 - 75	13%	9%	8%	10%	3%	7%	5%
76 - 100	2%	1%	3%	2%	2%	1%	4%
> 100	5%	2%	3%	5%	4%	6%	5%
Average lead* (µg/l)	28	21	22	25	16	19	22

IRON							
CONCENTRATION RANGE (µg/l)	OCT/NOV 87 NO DOSING	FEB 88 NO DOSING	MAY/JUNE 88 POLYPHOSPHATE	JUL 88 POLYPHOSPHATE	OCT 88 POLYPHOSPHATE	FEB 89 POLYPHOSPHATE	MAY/JUN 89 POLYPHOSPHATE
0 - 25	-	0%	18%	17%	3%	0%	1%
26 - 50	-	2%	58%	62%	64%	26%	48%
51 - 75	-	33%	15%	11%	22%	46%	37%
76 - 100	-	35%	3%	6%	7%	17%	8%
> 100	-	30%	6%	4%	4%	11%	6%
Average iron (µg/l)	-	101	50	44	49	74	58

* Average lead concentrations excluding lead concentrations > 100µg/l

Table 6 - Distribution of lead and iron concentrations during surveys in Area Fb

LEAD								
CONCENTRATION RANGE (µg/l)	JAN/FEB 88 NO DOSING	MAY 88 POLYPHOSPHATE	JULY 88 POLYPHOSPHATE	AUG 88 POLYPHOSPHATE	NOV 88 POLYPHOSPHATE	JAN 89 POLYPHOSPHATE	MAR 89 POLYPHOSPHATE	JUL 89 ORTHOPHOSPHATE
0 - 25	62%	64%	61%	67%	72%	69%	87%	69%
26 - 50	22%	22%	26%	17%	21%	19%	8%	22%
51 - 75	6%	4%	8%	4%	5%	4%	3%	4%
76 - 100	4%	5%	3%	4%	1%	4%	1%	1%
> 100	6%	5%	2%	8%	1%	4%	1%	4%
Average lead* (µg/l)	23	21	24	21	17	19	15	19
IRON								
CONCENTRATION RANGE (µg/l)	JAN/FEB 88 NO DOSING	MAY 88 POLYPHOSPHATE	JUL 88 POLYPHOSPHATE	AUG 88 POLYPHOSPHATE	NOV 88 POLYPHOSPHATE	JAN 89 POLYPHOSPHATE	MAR 89 POLYPHOSPHATE	JUL 89 ORTHOPHOSPHATE
0 - 25	0%	17%	4%	7%	22%	1%	1%	5%
26 - 50	10%	44%	78%	32%	59%	53%	78%	58%
51 - 75	66%	28%	15%	38%	11%	35%	13%	25%
76 - 100	15%	7%	2%	17%	4%	5%	3%	8%
> 100	9%	5%	1%	6%	4%	6%	5%	4%
Average iron (µg/l)	70	52	41	61	40	54	48	50

* Average lead concentrations excluding lead concentrations > 100µg/l

Table 7 - Results of statistical analysis of the effect of polyphosphate dosing on lead and iron concentrations in two sub-areas compared with the expected seasonal changes

DATE		EXPECTED CHANGE	MEASURED CHANGE			
			Area Fb		Area Fa	
		Lead and iron	lead	iron	lead	iron
No dose	Poly P					
Feb 88	May 88	increase	no change	decrease	no change	decrease
Feb 88	Jul 88	increase	no change	decrease	no change	decrease
Feb 88	Aug 88	increase	no change	decrease		
Feb 88	Oct 88	no change/increase			decrease	decrease
Feb 88	Nov 88	no change	decrease	decrease		
Feb 88	Jan 89	no change	decrease	decrease		
Feb 88	Feb 89	no change			no change	decrease
Feb 88	Mar 89	no change/increase	decrease	decrease		

4. CONSUMER SURVEY

During each sampling survey consumers were asked a questionnaire relating to the quality of their tapwater during the year prior to being questioned. The results are summarised in Table 8.

The main conclusions that can be drawn from these results shown in Table 8 are:

1. In general there were very few problems, except in Area F, and
2. Dosing polyphosphate in Area F significantly reduced the numbers of dissatisfied customers.

5. OVERALL CONCLUSIONS

Table 9 summarises the conclusions that can be drawn to date from the results of all the trials.

Table 8 - Summary of results from customer questionnaires

DOSE PHOSPHATE	DATE	AREA	NUMBER OF Q'AIRES	NUMBER OF PROBLEMS OF DISCOLOURED WATER OR BITS				
				TOTAL (%)	DAILY	WEEKLY	MONTHLY	YEARLY
Poly	July 88	Fa	210	26 (12)	1	6	9	10
Poly	March 89	Fa	100	8 (8)	4	2	2	0
Ortho	July 89	Fa	101	2 (2)	2	0	0	0
Poly	July 88	Fb	133	18 (13)	2	3	7	6
Poly	Jan 89	Fb	101	4 (4)	1	0	3	0
Poly	Feb/March 89	Fb	101	1 (1)	0	0	0	1
Ortho	June/July 89	Fb	100	0 (0)	0	0	0	0
-	Feb 89	E	101	2 (2)	1	1	0	0
Ortho	April/May 89	E	101	2 (2)	1	1	0	0
Ortho	June 89	E	101	0 (0)	0	0	0	0

Table 9 - Summary of remedial actions and effects

AREA	TYPE OF LEAD PROBLEM	TREATMENT	EFFECT ON AVERAGE Pb	EFFECT ON PEAKS Pb	EFFECT ON AVERAGE Fe	EFFECT ON PEAKS Fe	THEORY
E	Particulate lead	Ortho P dosing	Decrease	Expected seasonal increase does not occur	Decrease	Decrease	Ortho P is thought to affect lead through lowering iron conc. Mechanism unknown
D	Particulate and soluble lead	Kalipol Ortho P	No effect Decrease	No effect Decrease	No data probably not effective	No data probably not effective	Ortho P is thought to work on soluble fraction?
Fa/Fb	Particulate lead	Kalipol	Decrease Expected seasonal increase does not occur	No effect	Decrease	No effect?	Poly P works on iron concentrations and indirectly on lead concentrations
		Ortho P	Insuficient data.........				

APPENDIX E

FILTER SURVEYS

APPENDIX E

FILTER SURVEYS

E.1 Design of Surveys

Two areas known through previous work to have experienced problems with particulate lead have been identified within the UK. In each area, six properties have been identified, all still supplied by lead service pipes. On three of these properties, a filter of known pore size was to be fitted onto the communication pipe, as close as possible to the main. The other three properties act as controls.

All properties were to be sampled weekly at both low and high flow rates. Where sampling is going ahead, total and dissolved lead and iron are being analysed. The filter cartridges are being changed fortnightly to prevent clogging of the filter pores. Thirty weeks of sampling are intended, over the period July - December.

E.2 Filter Specifications

An in-line filter was required which could operate under the conditions prevailing in the service pipe and which would exclude particulate iron without otherwise affecting water quality or quantity. For these reasons a filter was selected with the following specification:

Cartridges:	1μm and 5μm pore size rating Constructed from pleated paper Maximum flow rate: 12 gallons/minute Maximum sustainable pressure: 125 psi.
Housing:	Manufactured from polypropylene with nitrile seals. (No metallic components.)

The filter cartridge was tested for its tendency to block and for its efficiency at excluding particulate iron. The results for the 1μm pore size filter are presented in Table E.1.

Table E.1 - Effect of filter (1µm pore size) on iron levels in tap water

DATE	TIME	BEFORE FILTERING (µg/l)			AFTER FILTERING (µg/l)			% REMOVAL		
		TOTAL IRON	DISSOLVED IRON	PART. IRON	TOTAL IRON	DISSOLVED IRON	PART. IRON	TOTAL IRON	DISSOLVED IRON	PART. IRON
12/6/90	am	80			80			0		
	pm	130			79			39		
13/6/90	am	196			133			32		
	pm	73			64			12		
14/6/90	am	95			63			34		
	pm	66			63			5		
15/6/90	am	100			64			36		
	pm	95			133			-40		
9/7/90	am	318	67	251	101	49	52	68	27	79
	pm	76	29	47	40	39	1	47	-34	98
10/7/90	am	90	59	37	56	43	13	42	27	65
	pm	61	61	0	47	41	6	23	33	-
11/7/90	am	51	46	5	51	37	14	0	20	-180
	pm	61	59	2	51	40	11	16	32	-450

Total iron removal was in the region of 30 - 50%. Of the particulate iron measured (ie the fraction exceeding 0.45µm in size) commonly 80 - 100% was excluded by the filter.

The removal of dissolved iron (<0.45µm) was probably due to adsorption onto the filter medium or onto particulates caught up in the filter.

Some problems were experienced due to poor sealing of the filter cartridge into its housing. Particulate material was allowed to pass through the filter on one day of the test (11/7/90).

From these results it was concluded that particulate iron-rich material would be sufficiently excluded from lead service pipes once the filter had been fitted.

E.3 Fitting of Filters

Properties were selected with short communication pipes and long lead supply pipes. The filters were mounted on 1" MDPE piping which then replaced a length of the communication pipe. The replacement was carried out with as little disturbance to the lead pipes as possible to minimise mechanical disruption of the corrosion products. The lead pipe cut-outs were retained for inspection.

E.4 Initial Results - Area B

Properties experiencing problems with lead were selected from the archive data. Random spot samples were taken at low and high flow to confirm the presence of lead. Results of lead and iron concentrations from the properties selected are given in Table E.2.

Table E.2 - Results of random daytime sampling - Area B

Property	Total lead µg/l	Total iron µg/l	Intended Control (C) or fitted with filter (F)
1	89 397	45 123	F
2	23 21	48 43	C
3	14 31	30 33	F
4	27 790	35 416	C
5	78 31	57 37	F
6	115 47	64 46	C

After the fitting of the 1µm pore size filters, random daytime samples were taken to establish that no significant mechanical disturbance to the lead levels had occurred. In general, lead levels were lower than those measured previously and are given in Table E.3.

Table E.3 - Results of random daytime sampling immediately following fitting of filters - Area B

Property	Total iron µg/l	Dissolved iron µg/l	Total lead µg/l	Dissolved lead µg/l
1	36	35	17	14.5
3	39	26	70	21
5	39	34	26.5	17

E.4 Initial Results - Area A

Properties experiencing problems with lead were selected from archive data and local knowledge. Potential properties and lead levels determined from overnight stagnation tap water samples are given in Table E.4.

Table E.4 - Results of overnight standing and 30 minute stagnation samples at selected properties - Area A

Property	Stagnation period	Total lead (µg/l)	Total iron (µg/l)	Total manganese (µg/l)	Filter to be fitted?
1	overnight	102	139	56	No
	30 mins	111	142	42	
2	overnight	8	206		Yes
	30 mins	11	64		
3	overnight	13	24		Not known
	30 mins	8	53		
4	overnight	19	145		Not known
	30 mins	24	153		
5	overnight	46	21		Not known
	30 mins	22	26		
6	overnight	Results not received			Not known
	30 mins				

A filter was installed at property 2 by by-passing the short side lead communication pipe with a new MDPE pipe to which the filter had been attached.

Initially a 1µm pore size filter cartridge was used but problems were experienced with clogging of the filter pores after one day. Consequently, a new filter cartridge was selected with a pore rating of 5µm. This too, blocked up after only two days in commission. High manganese levels were measured at property 2 not found in the other areas. It is possible that this may be contributing to filter blocking.

A 10µm, large surface area and a 20µm cartridge were investigated to see if they could be used.

Following these comprehensive initial investigations only properties 3 and 4 were found suitable for a long-term study. Trials then commenced using a 10µm filter.

APPENDIX F

RESULTS OF FIELD TRIALS INVESTIGATING REMEDIAL ACTIONS TO COMBAT THE OCCURRENCE OF FLAKING LEAD IN AREAS A AND B

APPENDIX F

F.1 Area A Study

The lead and iron data for the pair of sampled properties in Area A are presented in Table F.1. The dissolved lead and iron figures represent samples filtered through a 0.45µm pore membrane.

Samples collected from the two properties in Area A during the period January to August generally had low levels of lead.

The filtered property on one occasion showed a high soluble lead content of 75µg/l (95% of total lead) in a low flow sample, and a high particulate lead content of 89µg/l (90% of total) associated with high particulate iron in a high flow sample. This was the only occasion during this sampling exercise when particulate lead was monitored in substantial quantities.

Table F.1 - Summary of total and dissolved lead and iron concentrations for filtered and non-filtered properties in Area A

(period January to August)

PROPERTY	SAMPLE TYPE	NUMBER OF SAMPLES§	LEAD - TOTAL (µg/l)		LEAD - DISSOLVED (µg/l)		IRON - TOTAL (µg/l)		IRON - DISSOLVED (µg/l)	
			MEAN	MAXIMUM	MEAN	MAXIMUM	MEAN	MAXIMUM	MEAN	MAXIMUM
1. Control	High flow	31	10	43	>5	30	50	111	<20	35
	Low flow	31	9	16	11	183**	36	73	<20	28
1. Filtered	High flow	31	7	89	<5	15	44	151	23	53
	Low flow	31	6	79	7	75	31	80	<20	35

** Possible sample contamination (183 µg/l dissolved lead associated with total lead of 16µg/) otherwise maximum is 14µg/l and mean 6µg/l).

§ For iron dissolved N = 18.

F.2 Area B Study

The lead and iron data for each pair of properties in Area B are summarised in Table F.2. The dissolved lead and iron figures represent samples filtered through a 0.45µm pore membrane.A statistical analysis of all data has indicated the following.

(1) There is no consistent or significant difference between total lead in filtered and in non-filtered properties.Similarly for dissolved lead.

(2) Total iron is generally significantly lower in filtered premises. Dissolved iron is mostly not significantly different or is occasionally significantly lower in filtered premises. Thus the filters successfully remove particulate iron.

(3) Total lead is significantly higher in high flow samples than in low flow samples. Dissolved lead is either similar for both sample types or higher in high flow samples. Thus the difference in total lead levels between low and high flow samples is generally caused by more particulate lead in high flow samples.

(4) In high flow samples, total iron is higher than or similar to that in low flow samples. Dissolved iron is generally similar for both sample types. Thus where there is a difference in total iron levels between low and high flow samples, this is most likely to be caused by a greater amount of particulate iron in high flow samples.

(5) There is a significant correlation between total iron and total lead for both filtered and non-filtered premises.

Table F.2 - Summary of total and dissolved lead and iron concentrations for filtered and non-filtered properties in Area B (period July to December)

PROPERTY	SAMPLE TYPE	NUMBER OF SAMPLES	LEAD - TOTAL (µg/l)		LEAD - DISSOLVED (µg/l)		IRON - TOTAL (µg/l)		IRON - DISSOLVED (µg/l)	
			MEAN	MAXIMUM	MEAN	MAXIMUM	MEAN	MAXIMUM	MEAN	MAXIMUM
1. Control	High flow	22	200	1625	12	35.5	107	596	26	57
	Low flow	21	26	248.5	8	35	38	154	33	148
1. Filtered	High flow	22	93	1066	10	29	33	176	21	56
	Low flow	21	11	19.5	9	20	21	42	23	68
2. Control	High flow	22	24	48.5	12	25.5	41	80	24	52
	Low flow	22	16	33.5	12	49	37	75	27	55
2. Filtered	High flow	22	74	639.5	19	127.5	41	258	22	47
	Low flow	22	14	27	11	21.5	26	70	22	47
3. Control	High flow	22	192	2947	7	26.5	63	230	35	269
	Low flow	22	7	13	4	9.5	34	70	20	33
3. Filtered	High flow	22	818	7208	26	80	141	976	31	142
	Low flow	22	15	28.5	12	22	25	48	21	44

(6) No significant correlation could be found between temperature (between 4.5 and 24.5°C) and total lead and temperature (between 4.5 and 24.5°C) and total iron. However a significant seasonal effect (combined data from end of July to mid-September compared with combined data from end of September to early November) was found in total lead concentration in both filtered and non-filtered properties. A significant seasonal effect was found in total iron concentration in filtered premises but not in non-filtered premises. It is possible that part of this seasonal effect may be due to the destabilisation caused by mechanical disturbance during filter installation and the subsequent settling down; initial results were obtained during the summer period.

(7) Manganese and aluminium levels were generally low and are not likely to be linked with iron or lead in this area.

(8) The total alkalinity did not correlate with total lead or total iron. There was no marked variability in this parameter.

(9) The conductivity did not vary sufficiently to justify statistical analysis.

F.3 CO_2 Cleaning of service pipes

The results of this study are presented in Table F.3 and indicated the following.

Table F.3 - Summary of total and dissolved lead and iron concentrations for filtered properties and controls before and after CO_2 scouring of service pipe (filtered properties only) - Area B study

PROPERTY	SAMPLE TYPE	PERIOD*	NUMBER OF SAMPLES	LEAD - TOTAL (µg/l)			LEAD - DISSOLVED (µg/l)			IRON - TOTAL (µg/l)			IRON - DISSOLVED (µg/l)		
				MEAN	MAXIMUM	% > PCV	MEAN	MAXIMUM	% > PCV	MEAN	MAXIMUM	% > PCV	MEAN	MAXIMUM	% > PCV
2. Control	High flow	1	22	23	48.5	0	12	25.5	0	41	80	0	24	52	0
		2	35 (32)§	606 (61)	16050 (168)	20 (13)	16 (9)	134 (31)	6 (0)	230 (47)	3988 (313)	11 (3)	22 (21)	57 (57)	0 (0)
	Low flow	1	22	16	33.5	0	12	49	0	37	75	0	27	55	0
		2	35 (32)§	9 (8)	20 (20)	0 (0)	5 (5)	12 (12)	0 (0)	31 (31)	72 (72)	0 (0)	21 (22)	64 (64)	0 (0)
2. Filtered	High flow	1	22	74	639.5	27	19	127.5	5	41	258	5	22	47	0
		2	35	31	215	17	6	20.5	0	26	88	0	20	64	0
	Low flow	1	22	14	27	0	11	21.5	0	26	70	0	22	47	0
		2	35	4.5	12.5	0	3	14	0	22	79	0	18	62	0
4. Control	High flow	3	12	279	1125	83	7	14	0	111	254	17	25	128	0
		4	8	101	265	50	7	13	0	66	135	0	19	30	0
	Low flow	3	12	6	20	0	7	20	0	29	64	0	25	92	0
		4	8	10	56	13	5	28	0	29	57	0	22	42	0
4. Filtered	High Flow	3	12	801	5800	58	11	47	0	185	1146	17	20	57	0
		4	8	14	30	0	4	7	0	26	79	0	16	27	0
	Low flow	3	12	5	8	0	4	6	0	18	38	0	16	29	0
		4	8	4	8.5	0	5	13	0	14	21	0	21	37	0

* Period 1 - Before CO_2 scouring of filtered property (period July to December).
Period 2 - After CO_2 scouring of filtered property (period December to July).
Period 3 - Before CO_2 scouring of filtered property (period June to September).
Period 4 - After CO_2 scouring of filtered property (period September to November).
§ Figures in brackets exclude 3 samples taken when control premises were vacant.

(a) Property pair No.2

Lead and iron concentrations in low flow samples from both premises were low. There was no major effect of filtering on total and dissolved lead concentrations, although some evidence was obtained of an effect of filtering on particulate lead in low flow samples. The installation of the filter does have an effect on total iron both in low and high flow samples. The CO_2 scouring caused a significant reduction in lead in low flow samples (total, dissolved and particulate), although levels were already low.This effect did not reduce within the sampling period.This reduction was not observed in high flow samples.Similar peaks in concentration occurred both before and after the cleaning operation in both premises. The effect of the CO_2 scouring on iron levels is difficult to identify as total iron concentrations were always lower in the filtered premises.

Evidence from high flow samples indicates a positive and significant correlation between total lead and total iron, especially between particulate lead and particulate iron. There is a seasonal trend in total and dissolved lead concentration in low flow samples, concentrations being lower in the period after scouring, for both control and experimental properties. The high flow samples demonstrated a similar seasonal trend only in the experimental properties. It is of note that no significant effect was found for particulate lead. There was no obvious seasonal trend in iron concentrations.

For a 3-week period, the control property remained unoccupied. There was a dramatic effect of interruption in water demand on lead and iron levels of high flow samples. These reached high levels and were associated with elevated aluminium and manganese.

(b) Property pair No. 4

These premises were monitored for a shorter period than the previous properties. The results appear promising and are summarised below.

Lead and iron levels in low flow samples were very low in both properties, with no evidence of an effect of filtering or CO_2 scouring on lead concentrations. There is a significant decrease in total iron concentrations caused by filtering with possibly an additional effect of CO_2 scouring.

High flow samples from experimental and control premises show far more scatter and often show high peaks when the material is predominantly particulate. No effect of filtering on either lead or iron concentrations was evident in high flow samples. The CO_2 scouring of the service pipe effectively eliminates the particulate lead problem and causes a dramatic drop in particulate iron concentrations. The effect did not diminish during the 8 weeks of post-scour monitoring.

In high flow samples during the period when peaks in concentration occurred, there was a significant positive correlation between total iron and lead and between particulate iron and particulate lead.

No evidence of a seasonal effect on lead and iron concentrations was found for these premises.

These results for both pairs of properties do not provide consistent conclusions on the effect of CO_2 scouring. The property pair No. 4 provided promising results with particulate lead eliminated by the CO_2 scouring technique. The short post-scouring period does not permit the long-term effect of scouring to be established. The contrasting behaviour of property pair No. 2 may be a reflection of possible differences in procedure of the CO_2 scouring performed at each property.

The existence of such differences can however not be established.

APPENDIX G

INVESTIGATIONS ON EXHUMED SERVICE PIPES - ANALYTICAL TECHNIQUES AND RESULTS

APPENDIX G

G.1 Analytical techniques

G.1.1 Water samples

The total and dissolved (<0.45µm) iron, lead, aluminium and manganese levels were determined using inductively coupled plasma spectrometry; with a furnace atomic absorption spectrometry used to define lead levels if <50µg/l.

G.1.2 Pipe metal samples

The metal samples were digested using concentrated nitric acid (or aqua regia) and analysed using inductively coupled plasma spectrometry.

G.1.3 X-ray diffraction

The deposit samples were ground in a mortar and pestle and placed in a 0.3mm diameter amorphous glass tube in a Philips type PW/1024/20 X-ray camera. Cu Kα radiation was used with a nickel filter. Exposure time for each sample was 3 hours. For the sample from Area J, a 0.5mm tube was used as particles in this sample could not be finely ground.

Quantitative iron determinations were carried out on a Varian AA275 instrument using standard procedures, and X-ray fluorescence measurements were performed on a JEOL 35C Scanning Electron Microscope with a LINK AN 10 000 energy dispersive X-ray analysis system.

By recording the X-ray diffraction pattern of each of the powdered samples, and comparing the d-spacing of sets of lines comprising each pattern with those of known compounds by means of data in the Powder Diffraction File Search Manual, Hanawalt method (compiled by the JCPDS International Centre for Diffraction Data), it has been possible to identify compounds giving rise to the majority of the patterns. Confirmation was obtained by recording the X-ray powder patterns of authentic specimens of each compound so identified.

This technique will not identify non-crystalline organic constituents but can provide evidence of such components by their effect on the diffraction pattern obtained.

G.1.4 Microbiological plate counts and biofilm analysis

The following procedure was adopted for the microbiological inspection:

(i) To avoid contamination of the inside of the pipe all loose external soil deposits were removed. The ends of the pipe were sealed with a sterilised bung and the outside of the pipe was washed with tapwater.

(ii) The pipe was cut into 2.0cm sections, the first section was discarded.

(iii) To remove the adherent bacteria one end of the section was sealed with a sterile silicone bung and a volume (1.0 ml) of diluent (Ringers solution, pH 7.0) added. All adherent material was scraped from the pipe surface using a glass rod with a silicone tip. The "slurry" was made up to a final volume of 5.0 ml in more diluent. Vigorous shaking was used to produce a suspension of the slurry.

(iv) The bacterial population was determined from the number of colony forming units which appeared on R2A agar (7) using the spread plate technique, volumes (0.1 ml) of the appropriate dilutions of the slurry samples were spread on agar plates and incubated at 20°C for 7 days.

G.2 Results and conclusions

A summary of the results obtained is presented in Table G.1. The findings are discussed below.

G.2.1 Water quality at exhumation sites

The scale of the flaking lead problem in Area B was demonstrated by the fast flow samples from property 1. The extreme levels of particulate iron and lead were also associated with high levels of aluminium and manganese. At low flows lead and iron levels were low. Flaking lead was not visually apparent at property 2, although the analysis indicates a small quantity of particulate lead and iron.

The Area C tap samples contained little lead both at low and high flows. Iron levels were moderate with some present as particulate material at property 1 at high flow. Aluminium levels were moderate reflecting the aluminium sulphate coagulation at the corresponding treatment works; manganese levels were low. Area C tap water supplied by lead service pipes has historically contained particulate lead.

The low lead levels at the Area G property associated with soluble and particulate iron and low aluminium and manganese levels, did not reflect previous experience in this area where elevated total lead levels have been recorded. However, past experience shows that behaviour in the area can be variable and appears to be associated with certain properties or districts. The high TOC values of both sources to this area (Table 5.5) would be expected to promote particulate lead problems, compounded in one instance by elevated iron concentrations.

Table G.1 - Summary of water quality at exhumation sites and associated chemical and microbiological analyses

AREA	RANDOM DAYTIME (µg/l)		LOW FLOW (µg/l)				HIGH FLOW (µg/l)				PIPE METAL %	X-RAY DIFFRACTION INORGANIC ANALYSIS OF DEPOSITS	MICROBIOLOGICAL STUDIES CFU/1.0 CM PIPE LENGTH
			Pb		Fe		Pb		Fe				
	Pb TOT	Fe TOT	TOT	DISS	TOT	DISS	TOT	DISS	TOT	DISS			
C1	26	112	6	20	111	111	9	3	118	39	99.08 Pb, 0.90 Al, 0.02 Cu	$Pb_3(CO_3)_2(OH)_2$, SiO_2	38.6×10^3
C2	24	123	7	9	118	126	13	10	121	111	100 Pb	$Pb_3(CO_3)_2(OH)_2$, CuO	12.1×10^3
B1	36	31	8	4	16	20	1240) 15500)*	66	149) 1900)*	20	99.96 Pb, 0.04 Cu	Trace $Pb_3(CO_3)_2(OH)_2$ Trace SiO_2	1100×10^3
B2	9	29	3	8	22	27	26	3	136	26	99.85 Pb, 0.15 Zn	$Pb_3(CO_3)_2(OH)_2$, SiO_2	0.73 to 1160×10^3 Δ
G1	9	45	9	11	40	17	11	12	45	23	100 Pb	$Pb_3(CO_3)_2(OH)_2$	$21{,}000 \times 10^3$
G2	-	-	-	-	-	-	-	-	-	-	99.98 Pb, 0.02 Cu	Trace $Pb_3(CO_3)_2(OH)_2$ SiO_2	3025×10^3
H1	140	36	76	60	32	11	170	73	113	14	99.97 Pb, 0.03 Cu	$Pb_3(CO_3)_2(OH)_2$ (weak)+	665×10^3
H2	6	27	33	10	120	27	22	8	83	25	99.98 Pb, 0.02 Cu	+	1600×10^3
I	30	28	9	11	16	7	65	17	65	10	99.98 Pb, 0.02 Cu	$Pb_3(CO_3)_2(OH)_2$	978×10^3
J	14	59	4	3	16	7	22	4	20	14	99.98 Pb, 0.02 Cu	SiO_2	0.34×10^3

* Sample contained numerous flakes
Δ Non-uniform deposit, bacterial counts expressed as a range
\+ Mainly amorphous material

One Area H property registered high levels of particulate and soluble lead at high flows associated with a high particulate iron level. This property failed the lead PCV for all sample types. Lead levels at the second property were acceptable, although particulate iron was present. Aluminium and manganese were low.

Area I tap water, as represented by the single property, contained particulate lead in association with particulate iron at high flows. This contrasts with the recorded historical behaviour of a non-problem zone. Aluminium and manganese were low. The single property sampled in Area J contained acceptable levels of lead. This accords with the historical behaviour recorded for this area. Very little iron was recorded together with low aluminium and manganese concentrations. This area is supplied by spring sources which typically have high alkalinity and hardness and often low TOC, factors which make the conveyed water unlikely to cause particulate lead problems (see Section 2). The measured source alkalinity supports this (Table 5.5).

In summary, one property in Area B demonstrated clearly the typical flaking lead problem well documented in this area. Both Areas C and G gave no evidence of the soluble and elevated lead problems respectively encountered in the past. This serves to show the variable nature of lead problems and their non-appearance at certain properties or at certain times within problem zones.

Particulate lead was identified in Area H, where a soluble lead problem had been anticipated and also in Area I where historically no lead problem had been encountered. The association of particulate iron with the particulate lead confirms previous findings.

G.2.2 Pipe metal composition

All lead service pipes were almost or entirely 100% lead. A small quantity of aluminium (0.9% wt) and of zinc (0.15% wt) was detected in service pipes removed from Areas C and B respectively.

Traces of copper were present in 7 of the 10 pipes.

G.2.3 X-ray diffraction - inorganic analysis of corrosion deposits

A sample collected previously from Area A had already been examined using this technique.

A white material extracted from within an Area A pipe comprised lead carbonate hydroxide ($Pb_3(CO_3)_2(OH)_2$) as a predominant constituent and $PbCO_3$ at significantly lower concentrations. Other substances were also present in very low concentrations in this white material but could not be identified due to the low intensity of the X-ray lines. Dark grey particles, identified as lead, were also present in the deposit. Having a rough 'weathered' appearance it is assumed that these did not arise recently from sample extraction but existed in the corrosion layer in situ. Thus these results indicate lead to be the only metal present in substantial quantities.

As can be seen from Table G.1, basic lead carbonate ($Pb_3(CO_3)_2(OH)_2$) occurs in samples from Area C numbers 1 and 2, Area B number 2, Area G number 1 and Area I. It is also present in low concentrations (weak pattern) in sample Area H number 1, and it may also be present in trace amounts in samples Area B number 1 and Area G number 2, but the pattern was not complete or distinct enough to be certain. CuO (tenorite) was confirmed in sample Area C number 2, whilst silica, SiO_2, was present in samples Areas C number 1, B number 2 (and possibly number 1), G number 2 and Area J. Only in samples Areas C number 2 and B number 1, were sets of intense lines present for which a source could not be identified. A further investigation of these samples to attempt to identify these components is described below.

X-ray studies can only be used to identify crystalline materials, and the majority of samples appeared to contain some amorphous material. In particular the samples from Area H were almost entirely amorphous, and so their constitution could not be determined. X-ray fluorescence measurements were made on the samples from Area C number 2 and from Area B, in order to determine which elements were present, and with the following results:

Sample Area C Number 2: Cu, Ca, Al, Si, Fe, Mn, S and O
Sample Area B Number 2: Al, Zn, Si, C, S and O
Sample Area B Number 1: Fe, Ca, Al, Zn, Si, C, S, O and possibly F.

In addition, extraction of samples Areas G number 2 and H number 2 with acid followed by analysis by atomic absorption yielded a figure of ~ 8% for the iron content of both samples.

The theory of lead-water chemistry predicts that the corrosion deposits within a lead pipe will consist mainly of either or both normal lead carbonate ($Pb\ CO_3$) and basic lead carbonate ($Pb_3(CO_3)_2(OH)_2$). The carbonate type is determined predominantly by the pH and alkalinity of the water. The normal type occurs at low pH and the basic type at high pH; as alkalinity increases the pH for this stability transition increases. Under the conditions of alkalinity and pH existing for the sources, Table 5.5, basic lead carbonate is the predicted stable form. The X-ray diffraction studies have confirmed this prediction.

These results provide no consistent correlation between particulate lead problems and specific chemical components of the internal corrosion deposits. Basic lead carbonate was evident both at sites where particulate lead had occurred (e.g. Areas C and B), at sites previously demonstrating soluble lead problems (Area H) and in a non-problem area (Area I). However, sites in the last two categories were evidently experiencing minor levels of particulate lead during the current sampling programme.

Of note is the absence of lead-compounds in the material removed from the non-problem area J pipe. This suggests that the internal lead-containing deposits are very well adhered to the surface. The material collected from the bore surface for the X-ray examination representing the upper layer of deposit is likely to have been amorphous and therefore undetectable by this technique. The SiO_2 detected may have arisen from contamination of the pipe interior during exhumation.

The additional analysis using X-ray fluorescence identified a number of other constituent elements in the 3 samples examined (Areas C and B). The presence of other metals confirms previous findings.

G.2.4 Microbiological investigations

All of the pipes supported a bacterial population. However, with the exception of the Area G pipes, it is doubtful if the growth constituted a biofilm. A more appropriate description would be to consider that bacteria were associated with the adherent deposits, with localised colonisation or entrapment of bacteria within the deposit which appeared to be predominately non-biological in composition. In one of the Area B pipe samples, the bacterial count varied considerably along the length of the pipe; the highest count was associated with the sediment. The internal appearance of the Area G pipe samples suggested that these supported a biofilm. Certainly, the highest numbers of bacteria were recovered from these pipes.

The degree of colonisation as determined by the plate count technique did not correlate with the observed levels of particulate lead in the water samples collected prior to service pipe exhumation or with the historical behaviour associated with each of the areas. It is, however, possible that because the bacterial matter is associated with deposits in the service pipes, the microbiological character of the pipe is not stable and can fluctuate with conditions in the pipe, e.g. retention time, flow rate, etc., as these themselves affect the deposits. The conditions preceding pipe exhumation may therefore be important.

The pipe exhumed from an area supplied by ground water (Area I) supported much less microbiological activity.