EXPERIMENTAL STUDY OF LEAD CONTAMINATION IN DRINKING WATER SUPPLY SYSTEM OF HIGH-RISE BUILDINGS WITH LEAD-SOLDER JOINTS

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ABSTRACT

Lead (Pb) is a toxic heavy metal that may be present in household drinking water supply systems fitted with lead-containing fittings and fixtures. Worldwide there have been a number of excess lead incidents in drinking water supply systems, raising public health concerns. Even though lead pipes are not used in drinking water supply system of Hong Kong, excess lead has been found in the drinking water of high rise buildings of public rental housing (PRH) estates. It is suspected that the water is contaminated by the lead-solder joints and brass components in the supply chain. The objective of this study is to understand the lead release characteristics of a drinking water supply pipeline with lead-solder joints and brass fixtures under stagnant and flowing conditions. Leaching experiments in stagnant water are carried out using a brass valve and a lead solder joint connected with copper pipes. The results show that the mass of lead released from the brass valve and lead-soldered joint increases non-linearly with time and can be predicted using the Fickian diffusion law. Lead concentration measurements are conducted in lead-contaminated water supply chains consisting of components dismantled from (PRH) estates. High lead concentration is observed at the location of brass components and near locations of clusters of lead-solder joints. The lead concentrations of water supply chains with lead-solder joints are several times higher than that without lead-solder joints.

Keywords: Drinking water supply; lead contamination; lead solder; brass fixtures

1   INTRODUCTION

Drinking water is one of the main sources of lead (Pb) uptake by human beings. Long-term exposure to lead could result in learning deficits in children and increase blood pressure in adults, with children and pregnant women being the most vulnerable groups (WHO, 2011). The World Health Organization (WHO) has established a provisional guideline of 10 µg/L (ppb) as an upper limit for lead concentration in drinking water (WHO, 2011). In terms of health, the recommendation is to maintain lead levels in drinking water as low as reasonably practical. Worldwide there have been a number of widely publicized incidents of excess lead in drinking water, including Flint (Michigan, USA), Montreal (Canada) and New South Wales (Australia), and more recently in the high-rise rental public housing (PRH) estate buildings in Hong Kong, triggering great concerns among the public on the safety of drinking water.

In Hong Kong, a typical PRH estate building has 40 floors; each floor has about 20 flats/households. Treated water is first pumped to a roof top sump tank and then flows downwards along a downpipe connected to distribution system on each floor. The tap water supply for each flat is served by a single pipeline branched out from the downpipe in a common cubicle housing the water meters. The average length of water supply pipeline (from the meter cubicle to household tap) is about 10-30 meters. Due to the length and highly congested environment, a large number of soldered fittings (elbows, sockets and tees) are used to connect the copper pipes together, with fixtures (water meters, valves and water taps) made from brass materials. An extensive sampling program in PRH households has revealed that 57% of the "first-flush" samples (first sample of tap water in the morning) violate the WHO guideline (Lee, 2016). It is suspected that the drinking water is contaminated by lead released from lead-containing materials (leaded solder, brass fixtures) along the water supply chain and transported with flow to consumer's tap. The deposition, release and subsequent transportation of lead in such complicated labyrinth of water supplies in high-rise buildings is an emerging problem for many densely populated cities in the world that has not been previously studied.

Lead concentration of drinking water at consumer tap depends on a combination of factors including pipe material, fixtures and fitting material, water quality, the age of system and the water consumption pattern. Many works have been done on the lead contamination problems with lead pipes (Cartier et al., 2012; Del Toral, et al., 2013). While lead pipe is not used in drinking water supply systems in many countries including Hong Kong, the effect of lead-soldered connections and lead-containing fixtures becomes an increasing concern. Birden et

al. (1985) pointed out that the level of lead after 18-hour stagnation can be as high as 1 ppm in a newly constructed plumping system due to the lead-solder joints used. Kimbrough (2007) showed that brass components could be a major source of lead in drinking water by measuring lead contamination in all plastic pipes coupled by brass fixtures. Fig. 1 illustrates several hypothesized lead release mechanisms. The metal lead dissolves into water to form lead(II) (Pb^{2+}) ions. Lead dissolution is accelerated by electro-chemical (galvanic) coupling between lead solders and copper pipes, as the metal lead tends to lose electrons preferentially over copper (Oldfield, 1988). The lead ions diffuse into water, and are transported with the pipe flow. Soluble lead ions can combine with carbonate ions and hydroxide ions in tap water to form insoluble lead particulates which may deposit on the inner pipe surface in stagnation and mobilized under strong flows. Despite the evidence of lead leaching from lead solders and fixtures, there is hitherto no systematic study to quantify the lead release characteristics and transport under stagnant and flowing conditions.

The objective of this study is to understand the lead release characteristics of a drinking water supply pipeline with lead-solder joints and brass fixtures under stagnant and flowing conditions. Two series of experiments will be conducted to measure: (1) the lead release of a single lead-solder joint and a brass valve in stagnant water; and (2) the time variation of lead concentration in flowing condition after stagnation for pipe rigs resembling real-world water supply system with lead-solder joints and brass fixtures. This paper will start with the experimental details, followed by the results and discussions.
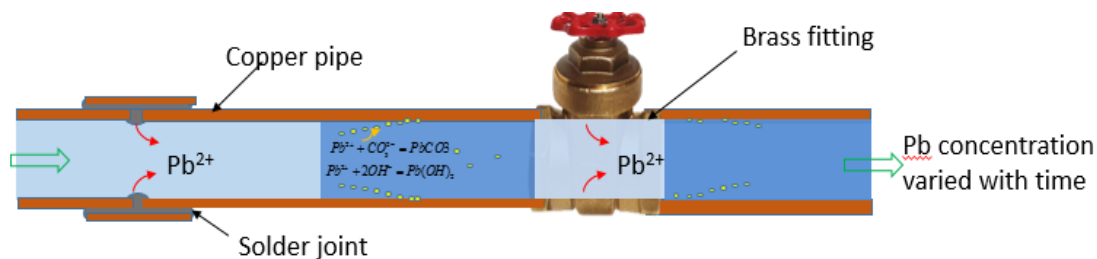

Figure 1. The release of lead (Pb) from galvanic corrosion and its transport in the water supply system with lead-soldered joints and brass fittings.

2   EXPERIMENTS

2.1 Measurement of lead release in stagnant water

In a drinking water supply system without lead pipe, the main sources of lead in the water are the leaded solder materials for joining pipes and the brass fixtures. The mass of released lead depends on complex factors: contacting area, contacting period, water quality, component material and chemical processes. The objective of the stagnation experiments is to measure the lead release characteristics of a lead-solder joint and a brass gate valve in stagnant water. The percentage of lead in lead solder material and brass valves are found to be 40-60% and 3-8% by weight respectively (WSD, 2015).

The experiment specimens are made with two 1.5-meter copper pipes connected by a lead-solder socket or a fully-opened brass valve. The internal diameter of the pipe is 20 mm, which makes the total volume inside the specimen is about 1 liter (Fig. 2). Before the experiment, the specimens are flushed with tap water for at least 5 minutes. Then the specimens are filled with tap water and sealed by rubber stoppers at two ends. Water stands still inside the specimens for a stagnation period from 0.5 to 18 hours. After the stagnant period, the water in each specimen is fully mixed and collected into a sample bottle. The experiments are carried out in room temperature (25ºC), and samples are stored in a refrigerator (below 4ºC) before lead measurement. The water samples are analyzed for lead concentration by an accredited lab (Health, Safety and Environmental Office, HSEO) in Hong Kong University of Science and Technology (HKUST) using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) method with a detection limit of 1 ppb. Selected experiments are carried out twice to ensure repeatability.

A 3m-long single copper pipe is used as a control to measure the background lead concentration in the tap water and any lead leaching from the copper pipe material at the same time. The lead concentration for all control experiments are lower than the detection limit of 1 ppb, suggesting there is negligible lead released from the copper pipe itself or in the fresh tap water.
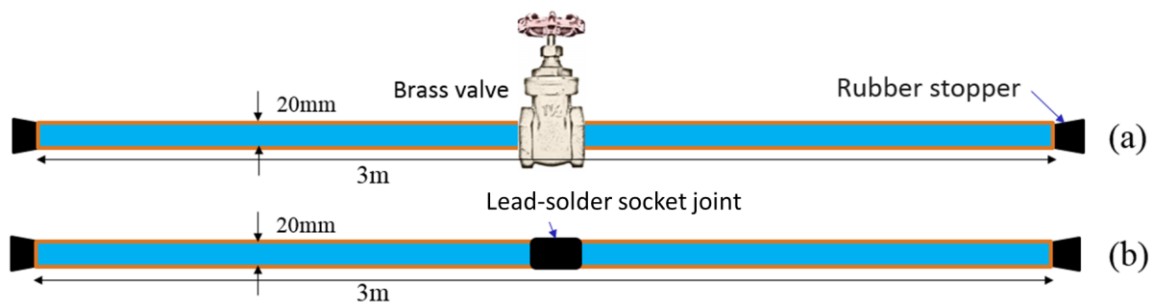
Figure 2. Specimens of lead leaching experiment in stagnant water, (a) copper pipes connected by a brass valve (fully opened); (b) copper pipes connected with a lead-soldered socket.

2.2 Measurement of lead concentration in lead-contaminated water supply chains

This series of experiments aims at understanding the lead release and transport in realistic household water supply chains and water consumption situations. The experiments are carried out at the electrical and mechanical workshop of the Water Supplies Department (WSD) of Hong Kong where assemblies of lead-contaminated water supply chains dismantled from public rental housing estates buildings are located. These water supply chains contains lead solder joints and lead-containing fixtures (brass valves, water meters, water taps with a goose neck and valve), resembling typical water supply pipelines in a typical PRH estate household with a length of 10-20 m (from the water meter to the kitchen tap).

A specifically designed experimental protocol is adopted to capture the time variation of lead concentration in the pipes. The pipe rigs are first flushed by tap water for 5 minutes with a fully opened tap. Then tap water is allowed to stay still inside the system for different period. At the end of stagnation, the tap is gradually opened to full. At the same time, two first-flush samples are collected by a 250 ml bottle and then a 750 ml bottle. Then three to four other 50 mL samples are collected subsequently at a time interval of 20-30 seconds. The flow rate is measured after each set of sampling. The water samples are collected carefully in order to avoid contamination due to the low level of lead (~10 ppb), and they are preserved in ice box immediately after collection before sending to accredited laboratories in HKUST and WSD for lead analysis by ICP-MS.

The experiments are conducted in one rig without lead-solder joints (Rig 4R) and two other rigs with leaded-solder joints (Rigs 3C & 2A). The layout and key components of rig 3C are illustrated in Fig.3 as an example. Rig 4R consists of only brass fixtures (brass valve, water meter and water tap), while more complicated route and more lead-containing components are present in Rig 3C and 2A. About 50 sets of experiments covering stagnation time of 0.5-68 hours are carried out with about 300 samples for lead concentration analysis. The characteristics of each pipe rig and the experiments are listed in Table 1.
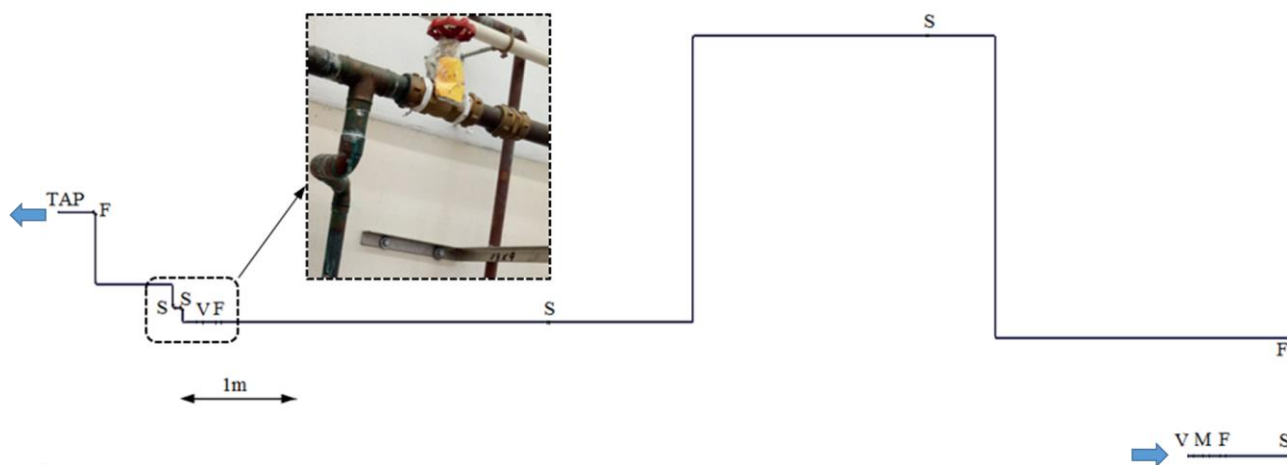

Figure 3. Layout of a typical drinking water supply line in a high-rise building (Rig 3C), with components dismantled from public housing estate of Hong Kong, V: valve, M: water meter, F: compression joint, S: lead-solder.

Table 1. Summary of experiments of measurement of lead concentration in water supply chains. Rig 4R has no lead-solder joints.

Test rig	4R	3C	2A
Pipe diameter (mm)	19.5	26	19.5
Pipe length (m)	10	18	16.5
No. of lead-solder joints	0	5	15
No. of brass components	4	8	9
No. of water samples	92	141	59
Flow rate range (ml/s)	30-120	37-110	100-176

3   RESULTS AND DISCUSSIONS

3.1   Lead release in stagnant water

Figure 4 shows the mass of lead (obtained by multiplying the lead concentration with total volume in the specimen ~1L) released from brass valves and lead-soldered joints, which increases non-linearly with time. The rate of lead leaching is the maximum at $t = 0$ and gradually decreases. After a stagnation period of 18 hours, the mass of lead released into water becomes stable and the leaching rates approaching zero. The leached mass of lead with a stagnation of 18 hours is with the same order as those reported in the investigation for exceed lead in drinking water in Hong Kong (WSD, 2015).

The diffusion of lead from the soldered joint and the brass valve is governed by molecular gradient diffusion and can be described by the Fickian diffusion law in cylindrical coordinates:

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial r^2} + \frac{1}{r}\frac{\partial C}{\partial r}\right)$$

where $C(x, r, t)$ is the lead concentration; D is the molecular diffusivity; t is time; x and r are axial and radial coordinates respectively. Immediately adjacent to the component wall, the dissolved lead concentration is in the saturated condition (Kuch and Wagner, 1983; van der Leer et al. 2002). A constant saturated lead concentration is prescribed at contact surface of the component with water as a boundary condition. The length of the brass valve L is 4cm and the internal contacting area with water can be estimated as 2.76×10^{-3} m^2 ($\pi \times d \times L$). For the soldered socket, it is assumed that the entire inner surface of the socket length (2cm) is covered with solder material and a contacting area of 1.38×10^{-3} m^2 is obtained. Based on the mass of lead released after 18 hours stagnation and the length of the component, the saturated concentrations of lead in water induced by a brass valve and a lead-soldered joint can be calculated as 284 and 2237 µg/L respectively. The above governing equation is solved numerically using a three dimensional (3D) grid using the Computational Fluid Dynamics (CFD) software FLUENT to obtain the spatial and temporal lead concentration variation with the molecular diffusivity of lead $D = 9.39 \times 10^{-10}$ m^2/s (Sato et al. 1996) and the initial condition of $C(x,r) = 0$ at $t = 0$. The prediction of the diffusion model agrees well with experimental measurements (Fig. 4).
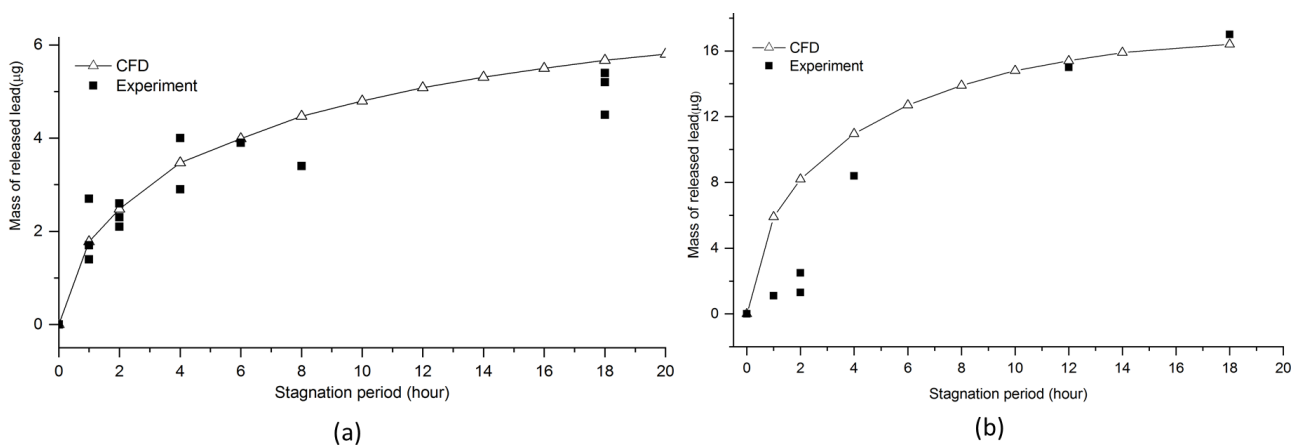

(a)   (b)

Figure 4. Mass of released lead and corresponding leaching rate variation with stagnation time of (a): brass valve and (b) lead-soldered joint. Mass of lead released in experiment and CFD model.

The distribution of lead in a cross section near a lead-solder joint after 2 and 18 hours stagnation is shown in Fig. 5, from which the diffusion process from the component surface to inner water body can be seen. At t = 0, a high lead concentration gradient exists between the water adjacent to the leaded components and those near the center of the pipe, thus there is a high diffusion flux of lead ions into the water. The concentration

gradient gradually decreases with time (Fig. 5a) and thus the increase in lead concentration gradually decreases. After 18 hours, the concentration in the water volume covered by the joint is close to the saturated concentration (Fig. 5b). The molecular diffusion is first completed in the radial direction (r) in the water volume within the component length. The diffusion in the axial (x) direction requires much longer time.
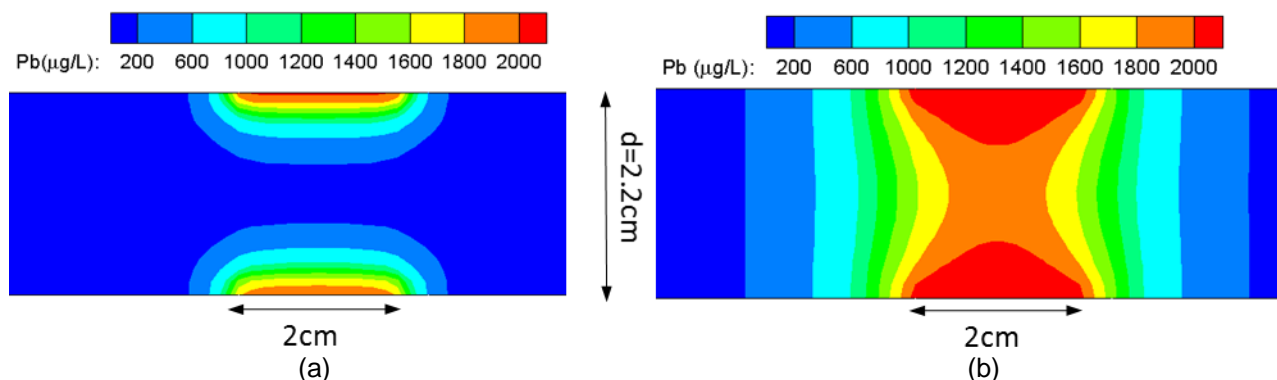

Figure 5. Predicted cross-sectional lead concentration distribution of a soldered socket joint (a): 2-hour stagnation, and (b) 18-hour stagnation.

3.2 Spatial variation of lead concentration in realistic water supply chains

The time variation of lead concentration at the tap in the "first flush" after stagnation depends on the location of lead sources, mass of lead leached during stagnation, the transport of lead by the flow and also possible pick-up of deposited lead particulates. Assuming the transport of lead is advection dominated, the time history can be transformed to spatial variation of lead concentration along the pipeline using a Galilean transformation $x = Ut$, where $U = Q/A$ is the average flow velocity; Q is the measured flow rate and A is the pipe cross-section area; t is the time after opening the tap. Locations of potential lead contamination in the pipeline can be traced using the longitudinal lead concentration profile.

Fig. 6a shows the lead concentration along the pipe length for a rig without any lead-soldered joints. Higher lead concentration is observed at the location of tap and a gate valve ($x = 0 - 200$ cm) and near the water meter and a brass gate valve at $x = 800 – 1000$ cm, while low lead concentration is observed for the pipeline itself. This indicates the water tap, meter, and valves made from brass materials are potential sources of lead contamination in the pipeline. The valves and meters could result in higher lead concentration, possibly due to the corrosion of lead from brass in the components and accumulation of lead particulates. The lead concentration in general increase with the stagnation time, but for the highest stagnation time of 68 hours, the lead concentration does not exceed the WHO provisional guideline of 10 ppb.

Fig. 6b shows the lead concentration for a pipe rig with lead-solder joints dismantled from PRH housing estates households. The peak lead concentration is found at the tap ($x = 0$), a cluster of lead-soldered joints ($x \sim 200$ cm) and at the cluster of a gate valve, a water meter and lead-soldered joints at $x = 1800$cm. The average lead level in this rig is several times higher than the pipe rig with only brass fixtures, consistent with the measurements in single-component stagnation tests. The lead concentration after 2 hours of stagnation can exceed the WHO provisional guideline and can be as high as 30 ppb.

Fig. 6c shows the lead concentration in a pipe with more leaded-solder joints (a total number of 15) all along the supply chain. The average concentration of tap water just after 0.5-hour stagnation time already exceeds the WHO provisional guideline. Peaks are observed at locations where intensive lead-solder joints exist, especially the elbow regions ($x = 1100$-1300cm). Lead concentration at elbow region can be as high as 65 ppb after 18 hours contacting period. In the elbow area, more solder materials are used and resulting in a larger contacting area for the solder with water. Lead particulates are also more easily to settle and accumulate at these positions of recirculating flows.

Besides being transported by the flow (advection), the leached lead is mixed over the pipe cross section by turbulence and the lead cloud will also be dispersed longitudinally due to the radial velocity shear, especially at recirculating zones of elbows and tee joints. Lead particulates may also be picked up by the flow and transported to the tap. A 3D flow and mass transport model is required to understand the mixing and transport of lead in such complex pipe system and correlate with the time history of lead level at the tap.
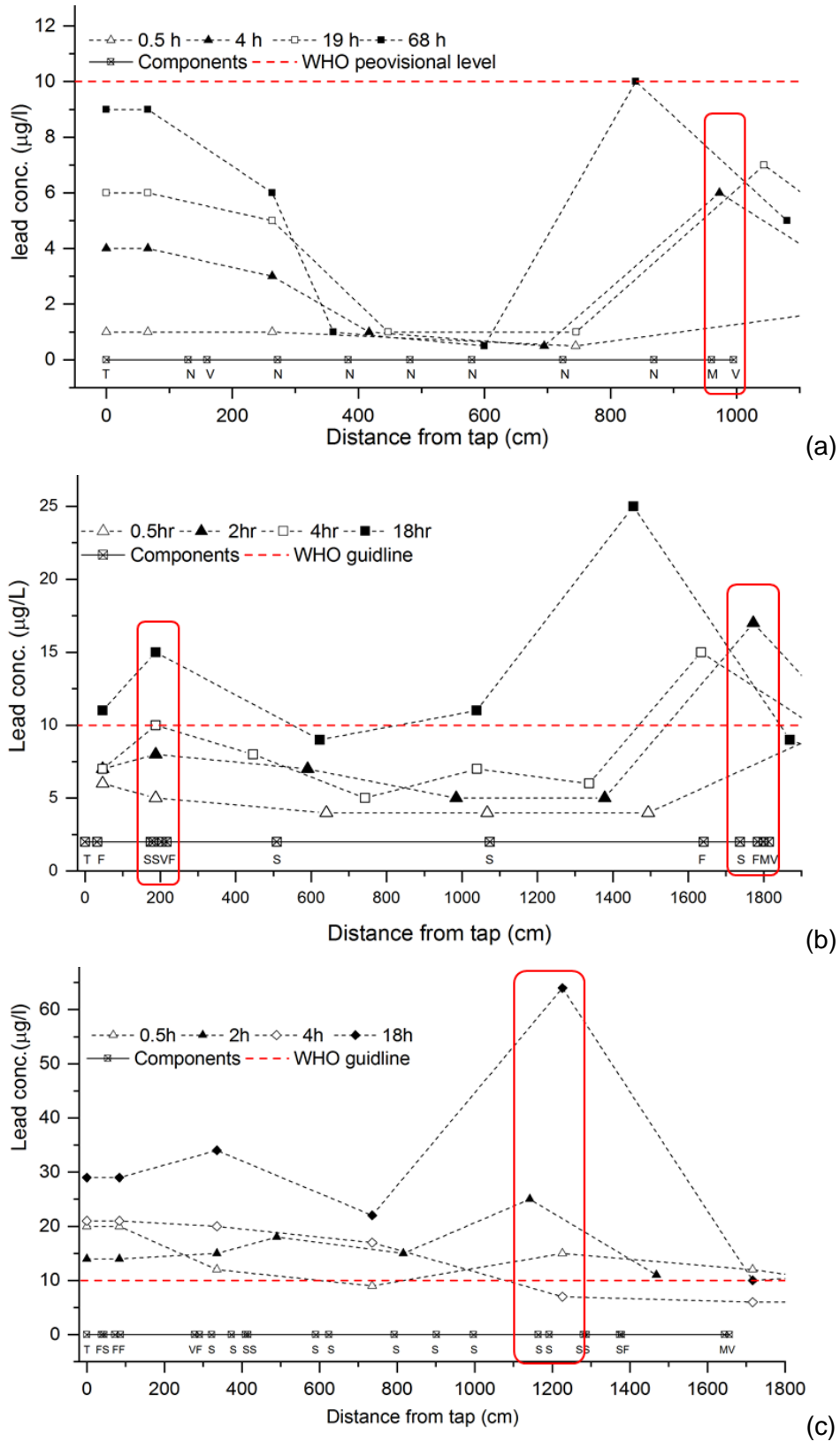
Figure 6. The spatial lead concentration distribution along (a) Rig 4R, (b) Rig 3C and (c) Rig 2A. The bottom line indicated the components on the water supply chain: S: lead-solder joint, F: compression joint, N: lead-free solder joint, T: tap, M: water meter.

4   CONCLUSIONS

This study shows brass components and lead-solders are the main sources of lead in drinking water supply systems without lead pipes. The mass of lead released from the brass valve and leaded solder increases non-linearly with time in stagnant water, but the leaching rate decrease exponentially with time. The lead leaching of lead solder joint is several times larger than that of brass material. Lead concentration time history at the water tap and spatial distribution in a typical water supply system in real life are obtained for the first time. The concentration peaks of lead concentration profiles along the pipeline fitted well with the locations of brass components or lead-soldered joints. The present experimental study provides a useful dataset for calibrating and validating a 3D CFD model for predicting the lead concentration variation at the tap to provide a basis for public health protection and water management.

ACKNOWLEDGEMENTS

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