## PREDICTION OF LEAD CONCENTRATION AT CONSUMER TAP OF LEAD CONTAMINATED WATER SUPPLY SYSTEM IN HIGH-RISE BUILDINGS

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## ABSTRACT

Lead (Pb) is a toxic metal that can be present in drinking water; exposure to lead (WHO standard C  $< 10 \mu g/l$ ) can result in learning deficits in children and increased blood pressure in adults. In 2015 a public outcry was caused by high excess lead concentrations found in the drinking water of public rental housing estates in Hong Kong. Similar incidents have been reported elsewhere.

Drinking water supply systems in densely populated high-rise buildings consist of complex labyrinths of copper piping fitted with solder joints and brass fixtures that contain lead. The prediction of lead concentration at the consumer tap is most challenging: the lead release mechanism due to galvanic corrosion and the interaction with the diffusion and transport in the pipe flow is largely unknown; the accurate measurement of the low concentrations of concern is also demanding and costly.

This paper presents a comprehensive experimental and theoretical investigation of lead contamination and transport in high rise buildings. Systematic tests on full-scale water supply chains dismantled from public rental housing estate buildings are performed. The main lead sources are determined; leaching experiments are performed on the key components to study the Pb leaching rate ( $R_t$ ) over time. A 3D diffusion model is developed to model the leaching process assuming a constant Pb concentration next to the pipe wall ( $E_0$ ), and the subsequent tap water concentration under flowing condition for a real-life water supply chain is successfully predicted by a numerical solution of the contaminant transport for the first time.

Keywords: Drinking water, lead contamination, lead-solder joint, equilibrium concentration, computational fluid dynamic model

# 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 increased 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  $10 \mu g/L$  as an upper limit for lead concentration in drinking water. Worldwide there have been a number of incidents of excess lead in drinking water, including Flint (Michigan, USA), Montreal (Canada), and more recently in the high-rise Public Rental Housing (PRH) estate buildings in Hong Kong, triggering great concerns among the public on the safety of drinking water.

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 (Chan & Lai, 2016). It is suspected that the drinking water is contaminated by lead released from lead-containing solders used in pipe connection. A typical PRH estate building in Hong Kong has 40 floors and 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 domestic plumbing system on each floor. The tap water supply for each flat is served by a single pipeline branched out from the down pipe 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 (Fig.1). Due to the length and highly congested environment, many soldered fittings (elbows, sockets and tees) and fixtures (water meters, valves and water taps) made from brass materials used to connect the copper pipes are possible lead sources. The size of these fittings and fixtures are small compared with the length of the piping; unlike lead pipes the scattered distribution of lead sources in the water supply chain presents additional modeling challenges.

Lead solder is used for pipe joints of water distribution systems in many countries; it can be a significant contributor of lead in potable water (Subramanian et al., 1991; Gregory, 1990). Lead-tin solder may present a particularly serious problem because of the galvanic nature of the solder-copper joint (Gregory, 1990; Subramanian et al., 1995; Reiber, 1991). Extremely high Pb concentrations (23.5 mg/L) have been observed next to the solder joint surface after 2h stagnation by micro-ion-selective electrodes (Ma et al., 2016, 2018); the local pH shift and chlorine consumption clearly demonstrate the occurrence of galvanic reaction. Copper alloy fixtures including brass and bronze may also lead to elevated lead at the consumer tap. Previous studies have shown the release of lead from inline brass fittings and fixtures can be significant – as high as 176  $\mu$ g/L at the tap (Dudi et al., 2005; Kimbrough, 2007; Elfland et al., 2010).

Chemical reactions, molecular and turbulent diffusion, shear dispersion and advection all play a role in determining the lead concentration at the consumer tap (Fig.2). The release of Pb from lead pipe under stagnant condition has been modeled by diffusion model and exponential model (Van der Leer et al., 2002). The equilibrium concentration ( $E_0$ ) at the wall of lead pipe has been determined for different water quality parameters and adopted in mass transfer model in flowing condition (Weatherill et al, 2000; Abokifa et al., 2017). However, the applicability of the equilibrium concentration approach for copper pipes with lead-containing fixtures and joints has not been studied. The relationship between the equilibrium concentration and galvanic corrosion processes has also not been examined. As far as we are aware, to date a comprehensive investigation aiming at the prediction of tap water lead concentration has not been reported.

In this paper, the equilibrium concentration at the surface of different lead-containing components are determined by calibrating a 3D diffusion model against experiment data of lead leaching experiments. Based on the calibrated  $E_0$  for different components, the time variation of lead concentration at the tap after different stagnation periods is predicted by CFD computation and compared with data.



Fig. 1 Water supply system inside a high-rise building of a Public Rental Housing estate of Hong Kong.



Fig. 2 Lead (Pb<sup>2+</sup>) due to galvanic corrosion and turbulent transport with flowing water to tap

## 2. 3D ADVECTION-DIFFUSION MODEL

#### 2.1 Mass-transfer model

In the macroscopic continuum approach a constant concentration  $(E_0)$  is assumed at the wall of lead-containing sources. Lead is then diffused from the source at the wall to the bulk water by molecular diffusion mainly during stagnation condition.  $E_0$  can be determined from leaching experiments after a long period of stagnation (usually overnight) in lead pipe (Van Der Leer et al., 2002). However, the experimental determination of equilibrium

concentration is more challenging due to the small length and unknown surface area of sources like solder joints. Alternatively, the equilibrium concentration can be determined from numerical modeling of leaching by calibration against experimental data. Once  $E_0$  of individual components is determined, it can be applied to a flowing water supply system.

### 2.2 Governing equation and numerical solution

The governing mass conservation equation in the water pipe flow can be written in terms of the lead concentration C(x, y, z, t) and the 3D fluid velocity field (u, v, w):

$$\frac{\partial C}{\partial t} + u \frac{\partial (C)}{\partial x} + v \frac{\partial (C)}{\partial y} + w \frac{\partial (C)}{\partial z} = (E_T + D) \left( \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right)$$
(1)

where  $D = 9.36 \times 10^{-10} m^2/s$  (Sato, 1996) and  $E_T$  are the molecular and turbulent diffusivities respectively. S and K are the sources and sinks of lead. The lead source is imposed at the boundary, not from the inner source term, the S term equal to zero. The concentration of lead in tap water is very low (parts per billion) and the simulation time is short (in the order of 1 minute). For the experiments of concern, the lead is mostly in the dissolved phase; the decay (deposition) is not considered. The numerical modeling proceeds in two steps: (i) for the leaching modeled by molecular diffusion in stagnation,  $E_T = 0$ , the lead source is prescribed by the equilibrium concentration at the pipe wall, and the time-dependent 3D diffusion equation is solved. (ii) After a period of stagnation, the concentration distribution resulting from the leaching from the wall is taken as the initial condition (or distributed sources) and the subsequent advective diffusion in the 3D flow is obtained by solving Eq. 2 using the CFD code ANSYS FLUENT. The turbulent quantities are modeled by two equations  $k - \varepsilon$  model. The turbulent viscosity under flowing conditions is determined locally as a function of k and  $\varepsilon$ ,  $\mu \sim k^2/\varepsilon$ , and  $E = \mu/Sc$ , assuming a turbulent Schmidt number.

The Dirichlet boundary condition (C=E<sub>0</sub>) is prescribed at the internal wall of solder joints and brass fixtures, and Neumann boundary condition ( $D \partial C / \partial z = 0$ ) is applied to the lead-free pipe walls. The initial concentration in the entire pipe is zero, and all the boundaries are set to be non-slip walls. When tap is open, a constant flow rate is prescribed at the inlet and pressure outlet boundary applied to the tap.

## 3. RESULTS AND DISCUSSION

## 3.1 Calibration of the equilibrium concentration from experiment data

Leaching experiments are performed in pipe specimen (inner diameter=26mm) with a solder joint or brass valve joining two 10 cm long copper pipes. The pipe is then filled with tap water with pH=8 and chlorine residual of 0.7 mg/L and left to stand for 0.5 to 24 hours. The full-mixing concentration at different times is measured (Fig.3). Corresponding leaching tests are also performed for copper pipe with lead deposits.

The measured length of the brass valve is 5cm, and the length of solder joint is assumed to be 1cm. Fig. 3 shows the comparison of predicted and measured lead concentrations during a leaching experiment with  $E_0 = 260$  and 1000 µg/L for the brass valve and solder joint respectively. The increase of concentration with time reflects a Fickian diffusion process, and in general the model is in good agreement with data. However, the model underpredicts the lead release at small times; this may reflect the inability of such a diffusion model to simulate the initial rapid release due to galvanic corrosion. Similar results are obtained with  $E_0 = 6\mu g/L$  prescribed at the entire inner surface of the pipe with depositions.



(a) solder joint

(b) brass valve

#### 3.2 Prediction of lead concentration at consumer tap

Applying the equilibrium concentration value ( $E_0$ ) of brass valve to all the brass components in a real life pipe system (Length=10m, D=20mm), the diffusion process from the four components are simulated first under stagnant condition. When the tap is turned on, the advection-diffusion equation is solved and the Pb concentration in the flowing water along the entire supply chain is computed (dashed line). To enable a meaningful comparison with the lead concentration of collected samples at the tap (1<sup>st</sup> 250 mL, 2<sup>nd</sup> 750 and 50 mL at t=30s, 50s,70s, 90s), the computed concentration at the tap is averaged according to the sample volumes. The volume-averaged concentration (solid line) at different times is plotted against the distance from the tap by a Galilean transformation (x=Ut) and shown against the different lead components along the pipe; it is seen that the model predictions compare well with the measurements (Fig.4). The four concentration peaks correspond to the four brass components (lead sources). For fixtures further away from the tap, the concentration is further reduced due to shear dispersion and turbulent mixing in the flow.



Fig. 4 Comparison of predicted and measured Pb concentration along a water supply chain; tap is turned on after 4- hour stagnation (Q=53ml/s, U=0.14m/s, Re=3800): T: tap, M: meter, V: valve.

Besides brass fixtures, Fig. 5 shows the comparison of predicted and measured concentrations for a real life supply chain dismantled from a high rise building (L=18m, D=26mm). The equilibrium concentrations are prescribed at the inner wall of solder joint (1000  $\mu$ g/L) and copper pipe with deposits (6 $\mu$ g/L). For this case, although there are multiple sources, only five peaks are observed at the tap – due to the merging, shear dispersion and cross-sectional mixing of sources. For example, the last peak in Fig. 5 is as high as 25  $\mu$ g/l and is due to five lead-bearing components (2 compression joints, a solder joint, a meter and a valve) – this implies that a sample at the water tap can be influenced by the lead sources 18 m away. The distributed release from deposits on the copper pipe can increase the background concentration in the entire pipe to 4  $\mu$ g/L and 6 $\mu$ g/L with 4h and 18h stagnation respectively.



Fig. 5 Comparison of predicted and measured Pb concentration along a water supply chain; tap is turned on after 4-hr stagnation (Q = 80 mL/s, U = 0.13 m/s, Re = 3345). T: tap, F: compression joint, O: solder joint, M: meter, V: valve.

#### 4. CONCLUDING REMARKS

An experimental and theoretical investigation has been carried out on the lead contamination and transport process in the water supply chain of high rise buildings. Based on the Fickian diffusion model, the equilibrium concentration at the wall of lead solder joints, brass fixtures and lead deposits are determined. It is shown that the Pb concentration at consumer tap of real-life water distribution systems can be successfully predicted by the equilibrium concentration approach. The results also show that the proportion of Pb from the deposits can exceed that from solder joints and brass components due to the distributed nature of the deposits. The connection of the advection-diffusion approach with galvanic corrosion is currently under study.

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