

Increases of lead and chromium in drinking water from using cement–mortar-lined pipes: initial modeling and assessment

Qizhong Guo*

*Department of Civil and Environmental Engineering, Rutgers, The State University of New Jersey,
P.O. Box 909, Piscataway, NJ 08855, USA*

Received 25 October 1996; accepted 5 March 1997

Abstract

A mathematical leaching model was developed for assessing potential increases of lead and chromium in drinking water from using cement–mortar-lined pipes. The initial wetting process, dissolution, diffusion, and advection of metals in the lining, accumulation of metals in the static pipe water, and dilution of metals by the flowing water were considered. Values for physical and chemical parameters in the model were obtained from literature or were estimated. Based on the modeled results, the drinking water standards are likely to be exceeded by using cement with lead solubility larger than 70 ppb or chromium solubility larger than 540 ppb. For one cement kiln studied, it is recommended that cement kiln dust not be fully recirculated when hazardous waste fuel (HWF) has the maximum allowable amount of lead. It is also recommended that the maximum allowable amount of chromium in HWF be lowered. © 1997 Elsevier Science B.V.

Keywords: Lead; Chromium; Drinking water; Cement–mortar lining; Water distribution pipe; Leaching model; Solubility; Porous medium; Hazardous waste fuel; Cement kiln

1. Introduction

Chromium in cement has been identified as causing human health problems by occupational dust exposure and skin contact. Lead builds up in the human body and is linked to damage to the brain, kidneys, nervous system and blood cells. The major source of lead in drinking water was identified to be plumbing materials, such as lead

* Corresponding author. Tel.: +1 908 445 4444; fax: +1 908 445 0577; e-mail: qguo@rci.rutgers.edu.

service pipes, solder, flux and fixtures. Amendments (1986) to the United States Safe Drinking Water Act (SDWA) prohibit the use of lead pipes. Also solder and flux that contain lead are prohibited under this act. Cement–mortar is a mixture of cement, sand, and water and is widely used in lining water distribution pipes. The cement–mortar lining is not considered a significant source of lead in drinking water. However, the recent practice of using hazardous waste fuel (HWF) in cement kiln has raised the question of whether much more lead and chromium (and other metals) will leach out from the cement–mortar lining to drinking water. An initial assessment of leachability of lead and chromium from cement–mortar lining and its implications for the practice of using HWF in cement kilns are made in this paper.

2. Elaborations on the necessity for an initial assessment

An initial assessment of potential increases of lead and chromium in drinking water from using cement–mortar-lined pipes is necessary for the reasons expounded below.

2.1. *Wide use of cement–mortar for lining drinking water pipes*

Of all pipes currently in use, 26% are cast-iron pipes and 17% are ductile iron pipes, which are already lined with cement–mortar. However, 22% are unlined, old cast-iron pipes [1]. Most old, unlined cast-iron pipes are corroded, resulting in reduced carrying capacity and problems with water quality. Cement–mortar lining is considered to be a cost-effective remedy for water-main corrosion [2–4]. A major effort is underway to line in-place corroded cast-iron pipes with cement–mortar, but this effort is largely undocumented in technical literature. If the unlined pipes are lined as part of a refurbishing effort, approximately 65% of the drinking water pipes in the United States will be lined with cement–mortar.

At this time, no restriction is imposed on the lead and chromium contents of cement for any use, including the lining of drinking water pipes [5]. Three standards were established by the United States water industry for cement–mortar-lined pipes [6–8]. However, none of them specify the limits of lead and chromium in cement–mortar linings.

2.2. *Elevated lead and chromium contents of cement because of using HWF in cement kilns*

More than 20% of cement kilns in the United States (23 out of 111) are routinely burning hazardous wastes [9,10]. Cement kilns in Europe and Canada also burn hazardous wastes. Burning HWF in cement kilns offers an alternative to disposal or incineration of certain waste streams [11]. This practice started about a decade ago and was intended mostly for decomposing hazardous organic compounds. However, some HWF contain not only toxic organic compounds but also toxic heavy metals. For a particular kiln investigated by Guo and Eckert [12], HWF contained 28 times more lead and 11 times more chromium than coal does for the same amount of thermal energy.

Heavy metals in fuels are not destroyed and may be discharged through three separate streams, including emissions from the kiln through exhaust stacks and incorporation into cement kiln dust (CKD) and clinker [12]. Clinker is grounded with a small amount of gypsum to produce commercial cement.

The cement industry collected and analyzed trace metal contents of clinker samples [13]. A total of 97 samples were taken from both HWF-burning and non-HWF-burning kilns. The mean chromium content for cement samples taken from kilns cofired with HWF was 113 mg/kg, which was statistically higher than that for cement samples taken from kilns burning coal, coke or natural gas (61.7 mg/kg). A similar significant trend was observed for chromium in CKD (56.8 vs 34.4 mg/kg). However, the mean lead contents for cement samples taken from HWF-burning and non-HWF-burning kilns were not statistically different (12.4 and 9.81 mg/kg, respectively). This is different from the significant trend for lead in CKD; the lead contents in CKD were 899 vs 250 mg/kg. The content of lead in cement will increase more significantly if CKD with a much higher lead content is recirculated to the kiln.

Limited data from the United States Environmental Protection Agency (USEPA) indicate increases in both lead and chromium contents of cement which was produced in kilns cofired with HWF [10]. During the USEPA's sampling program, 7 clinker samples were taken from kilns not using HWF and 9 from kilns using HWF. The average lead content of clinker from HWF-burning kilns was twice of that from non-HWF-burning kilns (9.3 vs 4.7 mg/kg). The average chromium content of cement from HWF-burning kilns was 37% higher than that from non-HWF-burning kilns (83.5 vs 60.9 mg/kg).

2.3. Increased leachable amounts of lead and chromium from cement produced in cement kilns cofired with HWF

The above reported contents of lead and chromium in cement were evaluated by using the USEPA's acid-soluble digestion method, method 3050 or 3051 in SW-846 [14]. One method assigned to evaluate the leachable amount of substances is the toxicity characteristic leaching procedure (TCLP), method 1311 in SW-846 [14]. From the cement industry's study [13], the mean chromium TCLP concentration of cement samples taken from kilns cofired with HWF was 623 $\mu\text{g}/\text{l}$, which was higher (but not statistically higher) than that of cement samples taken from non-HWF-burning kilns (497 $\mu\text{g}/\text{l}$). A similar significant trend was observed for chromium in CKD (145 vs 67 $\mu\text{g}/\text{l}$). However, the mean lead TCLP concentration of cement samples from HWF-burning kilns and non-HWF-burning kilns were not statistically different (6 and 7 $\mu\text{g}/\text{l}$, respectively). The mean lead TCLP concentrations for CKD were 114 vs 75 $\mu\text{g}/\text{l}$.

The USEPA's limited data indicate substantial increases in TCLP leachable amounts of lead and chromium from clinker that is produced in kilns cofired with HWF [10]. The average lead TCLP concentration for clinker samples taken from kilns burning HWF was 36 times that from kilns not burning HWF (250 vs 7 $\mu\text{g}/\text{l}$). The average chromium TCLP concentration for clinker samples taken from kilns burning HWF was about 3 times that from kilns not burning HWF (40 vs 15 $\mu\text{g}/\text{l}$). The USEPA has drawn no conclusions regarding the significance of any of the clinker data.

2.4. No comprehensive assessment yet of the leachability of lead and chromium from cement–mortar lining

Limited available data presented in Sections 2.2 and 2.3 indicate that both contents and leachable amounts of lead and chromium in cement were increased as a result of burning HWF in cement kilns. The leachability of toxic metals from cement–mortar cubes and concrete cylinders rather than cement powder was also tested [15–17]. In addition, a pilot laboratory study was conducted on the leachability of toxic metals directly from cement–mortar linings inside drinking water pipes [18]. A detectable amount of chromium, but a nondetectable amount of lead, was leached out from the cement–mortar cubes of Colucci et al. [15]. Detectable amounts of both lead and chromium were leached out from the concrete cylinders of Kanare and West [16], from the cement–mortar bars of Germaneau et al. [17], and from the cement–mortar linings of Guo et al. [18]. Although these laboratory results are indicative of the potential for lead and chromium to leach out from cement–mortar lining, they are limited in number and scope. As a result, they cannot be used to make an assessment on the leachability of lead and chromium under various leaching conditions, especially on whether the burning of HWF in cement kilns will cause excessive amounts of lead and chromium to leach out.

In this paper, a mathematical leaching model is developed as a tool for assessing the degree of increase in lead and chromium levels in drinking water from using cement–mortar-lined pipes. The governing equations in the model are solved analytically or by using numerical techniques. Projected values of lead and chromium concentrations will depend not only on governing equations and solution methods, but also on the values chosen for physical and chemical parameters in the equations. Therefore, a detailed literature search was made on the range of values for physical and chemical parameters. The conditions under which the action level for lead in drinking water ($15 \mu\text{g}/\text{l}$) and the maximum contaminant level of chromium ($100 \mu\text{g}/\text{l}$) would be exceeded are projected. The implications of these projections on the practice of HWF-burning in cement kilns are discussed. This initial assessment also provides guidance to future comprehensive laboratory and field studies.

3. Governing equations and solution methods for lead and chromium concentrations in pipe water

The concentration of metal (lead and chromium) in pipe water depends on the rate at which metal will leach out from cement–mortar lining and the dilution ratio in the pipe. The cement–mortar lining is a porous medium. Water has to seep into the lining first. Afterward, the rate at which metal will leach out depends on how much and how fast metal is dissolved and how fast metal will diffuse out from the pore water. A series of governing equations is derived for predicting the time variation of metal concentration in cement–mortar-lined pipes. A number of assumptions are made in deriving these equations; they are explicitly described.

A section of a circular pipe with a layer of uniform cement–mortar lining (Fig. 1) is considered. The cement–mortar-lined pipe has a certain length and an original inside diameter, D . The thickness of the cement–mortar lining is ΔR . Since the pipe is

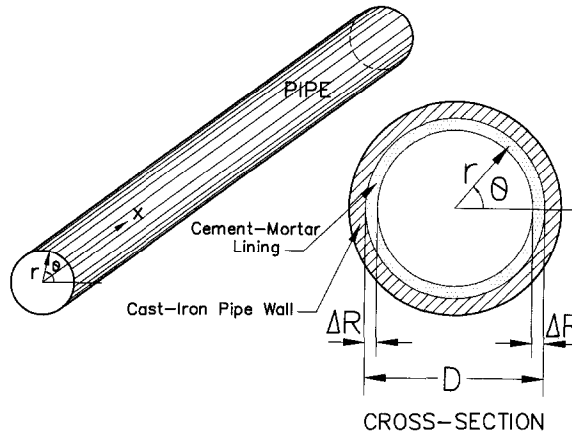


Fig. 1. Schematic of cement-mortar-lined pipe.

assumed to be circular and the thickness of lining is assumed to be uniform, all variables are axisymmetrical and do not change in the tangential (θ) direction.

3.1. Governing equation and solution method for water seepage velocity in cement-mortar lining

When the in-place cement-mortar-lined pipe is put into service, water starts to seep into the lining. A layer of coating material is applied on the surface of the cement-mortar lining in factory-lined pipes; this provides a barrier to the initial water seepage. However, there have been cases in which mortar coatings were cracked [19], which exposed the lining directly to water. In this assessment, only in-place lining is considered.

The Green-Ampt method [20] is used to derive the water seepage velocity. In the Green-Ampt method, a sharp wetting front is assumed (Fig. 2). At time t , the volume of water that has seeped into the lining is shown in Eq. (1):

$$F = \pi D \Delta R_w (\eta - \theta_i) \quad (1)$$

where F is the volume of water per unit length of the pipe that has seeped into the lining, D is the inside diameter of the pipe, ΔR_w is the distance at which the wetting front has moved into the lining, η is the porosity of the lining, and θ_i is the initial moisture content of the lining. The second order effect of ΔR_w on the circumference of the wetting front is neglected. From the conservation of mass, the water seepage velocity is shown in Eq. (2) and is the rate at which the amount of water seeps into the lining per unit area:

$$U_s = \frac{1}{\pi D} \frac{dF}{dt} \quad (2)$$

where U_s is the 'apparent' water seepage velocity, and t is the time. U_s is defined as the 'apparent' seepage velocity because it is the average velocity over the bulk area, including both pore and solid areas.

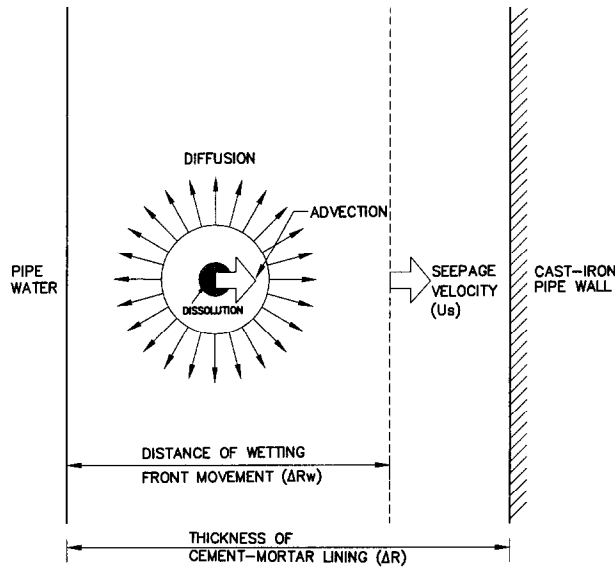


Fig. 2. Schematic of wetting front movement and transport processes in cement-mortar lining.

From the momentum point of view, Darcy's Law gives the 'apparent' water seepage velocity in Eq. (3) below.

$$U_s = -K \frac{dH}{dr} \quad (3)$$

where K is the hydraulic conductivity of the cement-mortar lining, and $\frac{dH}{dr}$ is the gradient of pressure head (H) in the radial (r) direction. If the pressure within the wetted zone is assumed to be linearly distributed, the water seepage velocity can be expressed as

$$U_s = K \frac{H_p + H_s}{\Delta R_w} \quad (4)$$

where H_p is the pressure head of the pipe water, and H_s is the suction head of the cement-mortar lining before the wetting.

Combining Eqs. (1), (2) and (4) gives

$$\frac{dF}{dt} = \frac{\pi^2 D^2 K (H_p + H_s) (\eta - \theta_i)}{F} \quad (5)$$

Integrating Eq. (5) and solving for F gives

$$F = \pi D \sqrt{2K(H_p + H_s)(\eta - \theta_i)t} \quad (6)$$

Based on Eqs. (2) and (6), the following 'apparent' water seepage velocity as a function of time is obtained:

$$U_s = \frac{1}{2} \sqrt{\frac{2K(H_p + H_s)(\eta - \theta_i)}{t}} \quad (7)$$

From Eqs. (4) and (7), the distance for which the wetting front has moved is:

$$\Delta R_w = \sqrt{\frac{2K(H_p + H_s)t}{\eta - \theta_i}} \quad (8)$$

After the wetting front has moved through the lining and has reached the cast-iron pipe wall, the seepage stops and U_s is equal to zero. Setting ΔR_w in Eq. (8) to be equal to the thickness of the lining (ΔR) gives T_s , the total time for the water to fill the pore space in the lining, as shown in Eq. (9):

$$T_s = \frac{\eta - \theta_i}{2K(H_p + H_s)} (\Delta R)^2 \quad (9)$$

3.2. Governing equation and solution method for concentrations of lead and chromium in pore water in cement–mortar lining

Based on the mass transport principle, metal (lead and chromium) concentration of the pore water in the cement–mortar matrix is governed by Eq. (10):

$$\eta \frac{\partial C_{\text{pore}}}{\partial t} + U_s \frac{\partial C_{\text{pore}}}{\partial r} = \frac{D_e \eta}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_{\text{pore}}}{\partial r} \right) - \rho_s (1 - \eta) \frac{\partial S}{\partial t} \quad (10)$$

where C_{pore} is the metal concentration of the pore water, η is the porosity of the cement–mortar, D_e is the effective diffusion coefficient, ρ_s is the density of cement–mortar based on the volume of solid, S is the metal content of the cement–mortar, r is the distance in the radial direction (Fig. 1), and t is the time. The first term on the left is the time variation of metal concentration in pore water at the location r inside the lining. The second term on the left is the rate at which the net amount of metal is advected into or out from the location r per unit volume (Fig. 2). The first term on the right is the rate at which the net amount of metal is diffused into or out of the location r per unit volume (Fig. 2). The second term on the right is the rate at which the amount of metal is generated or lost at location r per unit volume (Fig. 2).

If all the pores in the solid are connected and parallel, the effective diffusion coefficient can be related to the molecular diffusion (D_m), as shown in Eq. (11):

$$D_e = \frac{D_m}{\delta} \quad (11)$$

where δ is the tortuosity. The tortuosity is the ratio of the path length that a particle actually travels in the cement–mortar matrix between two locations to the direct geometric path length. Because the generation and loss of metal have already been considered in Eq. (10), incorporation of the chemical retardation into the effective diffusion coefficient is no longer necessary.

The amount of metal that will be released into the pore water from the solid matrix is assumed to be driven by the dissolution of metal compound inside the lining (Fig. 2). Under this assumption, the release rate of metal from the solid matrix to the pore water can be expressed as

$$\frac{\partial S}{\partial t} = -\alpha (C_s - C_{\text{pore}}) \frac{\eta}{\rho_s (1 - \eta)} \quad (12)$$

where α is the dissolution rate coefficient, and C_s is the solubility (saturation concentration) of the metal compound. Substituting Eq. (12) into Eq. (10) and rearranging the equation give:

$$\frac{\partial C_{\text{pore}}}{\partial t} + \frac{U_s}{\eta} \frac{\partial C_{\text{pore}}}{\partial r} = D_e \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_{\text{pore}}}{\partial r} \right) + \alpha (C_s - C_{\text{pore}}). \quad (13)$$

Eq. (13) for metal concentration in pore water is a time-dependent equation; thus, initial conditions (at $t = 0$) must be provided. From Eq. (13), metal concentration in pore water also changes in the radial direction (r direction); two boundary conditions must be provided to solve the second-order partial differential equation. At the interface of the pipe water and the cement–mortar lining ($r = D/2 - \Delta R$), metal concentration of pore water (C_{pore}) must be equal to that of pipe water (C_{pipe}). At the wetting front ($r = D/2 - \Delta R + \Delta R_w$) or at the interface of the cement–mortar lining and the cast-iron pipe wall ($r = D$), the diffusive flux (through the frontal cross section or the pipe wall) is set to be zero.

Eq. (13) is an advection–diffusion equation, even though the advection term will vanish after the wetting front reaches the cast-iron pipe wall. In addition, Eq. (13) has a source term (last term on the right-hand side). These characteristics of Eq. (13) render an analytical solution difficult, if not impossible. A numerical method is used to solve Eq. (13) instead. The explicit finite difference method [21] is used to represent the time derivative term in Eq. (13). The control volume method [22] is used to represent the transport (advection and diffusion) terms in Eq. (13). The upwind scheme is used to represent the advection term. The value of pipe water concentration at the previous time step is used as the boundary condition for the pore water at the lining–pipe-water interface. The value of the pore water concentration at the previous time step is used to determine the source term.

3.3. Governing equation and solution method for lead and chromium concentrations in the static pipe water

Static water may exist in the field at the dead ends of the water distribution pipe network or in the middle portion of the pipe network during the period of nonuse. In this case, metal (lead and chromium) that leaches out from the lining will accumulate in the pipe water.

The rate at which metal leaches out from the lining into the pipe water (J) is given in Eq. (14) and is equal to the rate of diffusion of metal out from the lining:

$$J = \pi (D - 2\Delta R) \eta D_e \left. \frac{\partial C_{\text{pore}}}{\partial r} \right|_{r = \frac{D}{2} - \Delta R} \quad (14)$$

where J is the net amount of metal which is leaching out from the lining surface per unit time per unit length of the pipe.

All leached metal will accumulate in the static pipe water. The increase of metal concentration with time in static pipe water is governed by the following equation:

$$\frac{1}{4} \pi (D - 2\Delta R)^2 \frac{dC_{\text{pipe}}}{dt} = \pi (D - 2\Delta R) \eta D_e \left. \frac{\partial C_{\text{pore}}}{\partial r} \right|_{r = \frac{D}{2} - \Delta R}. \quad (15)$$

Eq. (15) is derived assuming that the volume of pipe water is constant and assuming that metal concentration in the pipe water is uniform in the cross section and in the longitudinal direction. The first assumption is appropriate since the amount of water that initially seeps into the lining is very small in comparison to the total volume of pipe water and since an equal amount of water may be added to maintain the pipe pressure in practice. From Eq. (11), effective diffusion in the cement–mortar lining is a much slower process than molecular diffusion in static water because of physical retardation. Thus, metal that leaches out can be assumed to be fully mixed in the pipe water. Therefore, the second assumption is also appropriate.

Eqs. (7), (8), (13) and (15) are solved simultaneously to obtain the time variation of metal concentration in static pipe water. Euler's method [21] was used to solve Eq. (15). The metal concentration of pipe water is known at the initial time ($t = 0$). The metal concentration in the pore water at the time of initial wetting is assumed to be that of initial pipe water ($C_{\text{pore}0} = C_{\text{pipe}0}$).

Metal will continue to diffuse out to the pipe water until the metal concentration in the pipe water is equal to that of the pore water inside the lining or until the available amount of metal in the lining is depleted. The amount of metal that remains in each computational segment of the lining is checked at each computational time step. Once the originally available amount of metal at a location is found to be depleted, the dissolution rate is set to be zero at that location. Eq. (16) determines the total time duration (T_a) during which the available amount of metal in the entire lining will be depleted:

$$\int_0^{T_a} \left(\pi (D - 2\Delta R) \eta D_e \frac{\partial C_{\text{pore}}}{\partial r} \Big|_{r=\frac{D}{2}-\Delta R} \right) dt = \rho_s \pi (D - 2\Delta R) \eta C_{a \text{ mortar}} \quad (16)$$

where $C_{a \text{ mortar}}$ is the amount of metal in the cement–mortar lining available for leaching on the mass basis. Cement–mortar is made of cement, sand and water. After hydration, cement–mortar is a porous solid matrix. The availability of metal from cement–mortar can be calculated using Eq. (17):

$$C_{a \text{ mortar}} = \frac{C_a}{1 + R_{sc} + R_{wc}} \quad (17)$$

where C_a is the availability of metal based on the mass of cement, R_{sc} is the sand-to-cement ratio on the mass basis and R_{wc} is the water-to-cement ratio on the mass basis.

3.4. Governing equation and solution method for lead and chromium concentrations in the flowing pipe water

In the flowing pipe water, the time variation of metal (lead and chromium) concentration is affected by the rate at which metal leaches out from the lining, advection (dilution) by the incoming fresh water, and diffusion/dispersion in the pipe water. The

following mass transport equation is derived for the metal concentration in flowing pipe water:

$$\left(\frac{\partial C_{\text{pipe}}}{\partial t} + U_{\text{pipe}} \frac{\partial C_{\text{pipe}}}{\partial x} \right) \frac{1}{4} \pi (D - 2\Delta R)^2 = \pi (D - 2\Delta R) \eta D_e \left. \frac{\partial C_{\text{pore}}}{\partial r} \right|_{r = \frac{D}{2} - \Delta R} \quad (18)$$

where C_{pipe} is the metal concentration in flowing pipe water, U_{pipe} is the water flow velocity, x is the distance along the pipe (Fig. 1), and t is the time. Eq. (18) is derived assuming the metal concentration in the pipe water is uniform in the cross section (radial and tangential directions). This assumption is appropriate since the turbulence of flowing water will mix the leached metal fully in the pipe cross-section. Diffusion/dispersion in the longitudinal direction is omitted since it is usually much smaller than the advection term in pipe and river flows [23].

Eqs. (7), (8), (13) and (18) are solved simultaneously to obtain the metal concentration in flowing pipe water. The methods of solution to Eqs. (7), (8) and (15) are the same as those described in Section 3.3. The explicit finite difference method [21] was used to solve Eq. (18). To solve Eq. (18), the initial value of the metal concentration in the pipe water must be provided. At the upstream end of the pipe ($x = 0$), C_{pipe} is set equal to the known metal concentration of incoming water ($C_{\text{pipe}0}$).

3.5. Comparison with existing leaching models for similar environments

No model has been developed for assessing the leaching of lead, chromium, or other metals from cement–mortar linings inside drinking water pipes. However, several models have been developed for evaluating constituent leaching in similar environments. Leaching models have been applied to project leaching of constituents from solidified waste forms [24–26]. Kosson et al. [27] used a leaching model to project the released amounts of constituents from applications of municipal waste combustion (MWC) residue. All these models used diffusion equations. Analytical solutions for diffusion under various boundary conditions can be found in the work by Crank [28]. However, those analytical solutions cannot be applied directly to the cement–mortar lining case (Eq. (13)) because of the existence of both advection and source terms.

A leaching phenomena was observed in leaching cement-based waste forms by Cheng and Bishop [25] and Cheng et al. [29]. A clear-cut leaching boundary of approximately 100 μm in width was observed in every leached sample. The free hydrogen ions diffused into the solid matrix and neutralized the alkalinity provided by the binder in the leach front. The metals that precipitated previously in the high-pH environment are dissolved again and diffused outward into the leachant. A friable, silica-rich leached layer is formed and moves deeper into the solid matrix with time. This sharp leaching front, if it exists, will follow the wetting front described in Sections 3.1, 3.2, 4.2 and 5.1. However, in this study's case, the pH's of pipe water and pore water are normally high, and such an abrupt leach front is not expected to form. Seveque et al. [26] divided the dissolution process into an instantaneous process and a kinetically controlled process. If the dissolution is assumed to occur instantaneously, a dissolution front is formed, and the amount of element dissolved and the position of the dissolution

front versus time are controlled by the rate at which the dissolved element is diffused out. If the dissolution is kinetically controlled, the rate of dissolution is determined by the difference between the solubility (saturation concentration) and the actual concentration in the pore space and by the reaction rate, as expressed by Eq. (12). In this paper, diffusion and dissolution processes are considered simultaneously, and a gradual, instead of an abrupt leaching (dissolution) front is modeled.

4. Physical and chemical parameters in governing equations

The values of the physical and chemical parameters that must be specified to obtain quantitative solutions to the governing equations are listed in Table 1. They are described below.

4.1. Parameters for pipe geometry and flow (D , ΔR , water stagnation time and U_{pipe})

The two most common pipe network configurations used are the branching system and the grid system [30]. The structure of the branching system is similar to a tree. The trunk line is the main source of water supply. Service mains are connected to the trunk line, and submains are connected to the service mains. In turn, building connections used to provide service to individual residences and buildings are connected to the submains. The distinguishing feature of the grid system is that all of the pipes are interconnected, and there are no dead ends. In such a system, water can reach a given point of withdrawal from several directions.

The smallest inside diameter of branching pipes (D) is 200 mm (8 in.) [30]. The smallest pipes in a grid system are 150 mm (6 in.) in diameter. The thickness of the in-place lining (ΔR) depends on the diameter of the pipe and the pipe material [7]. For example, for old and new cast-iron and ductile-iron pipes of diameter 100 mm to 250 mm, the thickness of the lining is normally 3.2 mm (1/8 in.). For badly deteriorated pipes, a greater thickness may be used based on engineering judgment. In all instances, tolerance for lining thickness shall be +3.2 mm (+1/8 in.) with no minus tolerance.

The in-place lining procedure is mostly applied for rehabilitating old corroded pipes, which were commonly installed in a branching configuration. In a branching system, the condition of water stagnation at the dead ends could persist for a long period of time (i.e., one or more weeks). Under stagnation conditions (at the dead ends or during a period of nonuse), velocity is zero. Velocities of water in the middle portion of the pipe (U_{pipe}), during periods of use, are commonly between 0.6 and 1.25 m/s (2 and 4 ft/s) [30].

4.2. Parameters for calculating the movement of the wetting front (H_p , K , η , H_s and θ_i)

For typical residential rates of water demand, a static pressure of 275 kPa (40 lb/in², 28.0 m head) is considered to be normal [30]. The minimum recommended pressure is about 140 kPa (20 lb/in², 14.3 m head). In business districts, pressure values in the range of 350 to 550 kPa (50 to 80 lb/in², 35.7 to 56.1 m head) are common. The pressure head in the pipe (H_p) is equal to the pressure divided by the specific weight of water (9.81 kN/m³).

Table 1
Range of values for physical and chemical parameters

Symbols	Range of values	References
D	≥ 150 mm	Ref. [30]
ΔR	3.2 to 6.4 mm	Ref. [6]
None (water stagnation time)	Estimated to be from 8 h to 2 weeks	This study
U_{pipe}	0.6 to 1.25 m/s	Ref. [30]
H_p	14.3 to 57.2 m (20 to 80 psi)	Ref. [30]
K	1.0×10^{-13} to 1.0×10^{-10} m/s	Ref. [31]
η	0.19 to 0.48	Ref. [32]
H_s	Expected to be larger than that for clay (0.32 m)	Ref. [20] for clay
θ_1	Assumed to be zero	This study
C_a	Expected to be equal or smaller than acid-soluble content, which was measured to be from 0.13 to 75 mg/kg for lead, and 4.8 to 422 mg/kg for chromium. The projected maximum acid-soluble content of lead is 530 mg/kg for one cement kiln.	Refs. [10,13,35] for measured acid-soluble contents; this study for the projected maximum lead content
R_{sc}	2.0 to 3.0 derived from the volume-basis ratio of 1.0 to 1.5	Ref. [6] for volume ratio
R_{wc}	0.4 to 0.6	Ref. [6]
ρ_s	Estimated to be 2000 kg/m	This study
C_s	Assumed to be 1 to 4 times that of TCLP concentration at the same pH level. Measured TCLP concentrations of lead range from nondetectable to 1900 $\mu\text{g/l}$, and those of chromium range from 14 to 1540 $\mu\text{g/l}$. Final pH of TCLP fluid is around 12.5, similar to the pH of pore water in lining. Solubility of lead in drinking water at high pH is about 70 $\mu\text{g/l}$.	Refs. [10,13,35] for TCLP concentrations; Ref. [37] for lead solubility in drinking water
α	Estimated from TCLP and acid-digestion test protocols to be from 0.428 to 50 h^{-1}	Ref. [14] for test protocols
D_m	0.945×10^{-9} m^2/s for lead, 0.595×10^{-9} m^2/s for chromium	Ref. [39]
δ	Expected to be larger than that for concrete and smaller than that for cement paste. 200 to 400 for concrete. 510 for newly hydrated cement paste, 300 at 10% lime leaching, and 12 at 100% lime leaching	
$C_{\text{pipe } 0}$	Ref. [40] for concrete; Ref. [41] for cement paste Less than 15 $\mu\text{g/l}$ for lead, and less than 100 $\mu\text{g/l}$ for chromium less than 100 $\mu\text{g/l}$ for chromium	USEPA drinking standards [42]

Halamickova et al. [31] measured water permeability (k) in portland cement–mortars. They related the permeability to sand content, the degree of hydration and the water-to-cement ($w:c$) ratio. For sand content of 55% and $w:c$ ratio = 0.5, permeability decreases from 10^{-17} m² at 50% hydration to $10^{-18.5}$ m² at 70% hydration. For sand content of 45% and $w:c$ ratio = 0.4, permeability decreases from 10^{-18} m² at 50% hydration to 10^{-20} m² at 70% hydration. The relationship between hydraulic conductivity and permeability is shown in Eq. (19):

$$K = k \frac{\gamma}{\mu} \quad (19)$$

where K is the hydraulic conductivity, k is the permeability, γ is the specific weight of water and μ is the viscosity of water. Using the specific weight of 10 kN/m³ and viscosity of 1.0×10^{-3} Pa/s, the permeability of 10^{-17} m² is equivalent to the hydraulic conductivity of 1.0×10^{-10} m/s, and the permeability of 10^{-20} m² is equivalent to the hydraulic conductivity of 1.0×10^{-13} m/s.

Bourdette et al. [32] used models and mercury intrusion porosimetry data to calculate the porosity (η) of the bulk paste and the aggregate bulk paste transitional zone in cement–mortar. For a cement–mortar cylinder with $w:c$ ratio = 0.4, sand content of 52% and curing time of 3 months, the porosity of bulk paste was calculated to be 0.19, and that of the transitional zone was 0.48.

The suction pressure head (H_s) inside the porous cement–mortar before the wetting is unknown. Studies have been done on the dependence of soil suction head on soil type [20]. For clay, the suction head ranges from 0.064 to 1.565 m with an average value of 0.316 m. The suction head inside the cement–mortar is expected to be larger than that of clay at the same initial moisture content. The initial moisture content in cement–mortar lining (θ_i) is unknown. It is assumed to be very small in this study.

4.3. Parameters for leaching (C_a , R_{sc} , R_{wc} , ρ_s , C_s , α , D_m , δ , β , C_{pipe0})

The availability (C_a) is defined as the maximum quantity of soluble fraction of a residue constituent that can be released into solution under aggressive leaching conditions [27,33]. One standardized leaching procedure which results in the measurement of constituent availability is NEN 7341 [33]. In the availability leach test (ALT) applied by Kosson et al. [34], two serial extractions were carried out using distilled water, each with a 100:1 liquid-to-solid ($l:s$) ratio. The pH is kept at 7 during the first extraction and at pH 4 during the second extraction by using an automatic pH controller that delivers diluted nitric acid. Thus, the amount of acid added to the extraction fluid varies in order to maintain a constant pH level. The first and second extraction fluids are combined for analysis. A very large $l:s$ ratio ensures that the contaminant release is not constrained by the solubility. ALT is similar to NEN 7341, and it generally extracts all species not tightly bound in a mineral or glassy matrix.

Since ALT has not been applied for cement samples, measured acid-soluble contents are used as the availability in this paper. In the USEPA's acid-soluble digestion method (method 3050 or 3051), a representative sample of up to 0.5 g is digested in 10 ml of concentrated nitric acid for 6 min. It is expected that the acid-digestion method will yield more lead and chromium than the ALT method because of the stronger acid used,

unless the smaller $l:s$ ratio ($\leq 20:1$) in the acid-digestion method results in the solubility-constraint. From all measurements available, acid-soluble lead contents of cement ranged from 0.13 to 75 mg/kg, and acid-soluble chromium contents ranged from 4.8 to 422 mg/kg [10,13,35].

According to the lining standard [6], the approximate proportions of cement and sand in the mortar for the lining should be 1 part of portland cement to 1–1.5 parts of sand by volume. This is equivalent to the sand-to-cement ratio (R_{sc}) from 2 to 3 on the mass basis, if sand is assumed to be twice as heavy as cement. The water-to-cement ratio (R_{wc}) is normally 0.4 to 0.7 [6]. The solid density of mortar (ρ_s) is estimated to be 2000 kg/m³.

How much and how fast available metal will leave cement grains and enter pore water are assumed to be dissolution-controlled, as defined by Eq. (13). The acid neutralization capacity (ANC) test [36] was recommended to be used for measuring solubility (C_s) as a function of pH [27]. The solubility of most heavy metals is strongly a function of solution equilibrium pH. Solubility of a particular element can also be increased by the presence of significant concentrations of complexing agents such as chloride or acetate, or reduced by the presence of coprecipitating species such as sulfate or sulfide [27]. Solubilities of lead and chromium in the pore water of the cement–mortar lining and in the cement–mortar-lined pipe water have not been measured.

Solubility of lead in MWC residue was measured by Kosson et al. [27]. In their studies, 11 separate extractions were performed using separate samples at a $l:s$ ratio of less than or equal to 5:1. The low $l:s$ ratio results in the extraction being solubility-constrained for some analytes. Each extraction receives a different amount of diluted nitric acid, varying from 0 to 10 mg/g of dry residue, resulting in a range of final pH's. They obtained the lead solubility in MWC bottom ash of 40,000 $\mu\text{g/l}$ at pH 3; 10,000 $\mu\text{g/l}$ at pH 4; 70 $\mu\text{g/l}$ at pH 7; 15 $\mu\text{g/l}$ (minimum) at pH 9.3; 40 $\mu\text{g/l}$ at pH 11; 200 $\mu\text{g/l}$ at pH 12; and 1,000 $\mu\text{g/l}$ at pH 12.5.

Schock [37] predicted the solubility of Pb^{2+} in drinking water with the presence of hydroxide and carbonate species. The pH of water in newly lined pipes has been observed to be high because of the release (corrosion) of calcium hydroxide from the lining [38]. An even higher pH level is expected for the pore water inside the lining. Under high pH, the solubility of Pb^{2+} was predicted by Schock [37] to be about 70 $\mu\text{g/l}$.

If the pore water inside the lining has the same characteristics (e.g., pH and $l:s$ ratio) as the TCLP extraction fluid and if the metal content in the TCLP extraction fluid reaches the upper limit, the TCLP concentration can be used as the solubility of metal in the pore water. TCLP was applied to measure leachable amounts of metals from 16 cement samples by Eckert and Guo [35]. The extraction fluid 2 (diluted glacial acetic acid) with pH of 2.88 ± 0.05 was used, and the $l:s$ ratio was 20:1. Although the initial pH was low (2.88 ± 0.05), the final pH's for the 16 samples were from 12.22 to 12.69 (average at about 12.5). A similar pH level is expected in the pore water inside the lining. Since TCLP has a higher $l:s$ ratio (20:1) than that of ANC test (5 or less to 1), the solubility limit may not have been reached during the TCLP extraction. Therefore, the solubilities of lead and chromium in the pore water could be from 1 to 4 or more than that of the TCLP concentrations. The highest TCLP concentration obtained for lead

by Eckert and Guo [35] was $17 \mu\text{g/l}$, the highest by the Portland Cement Association (PCA) [13] was $29 \mu\text{g/l}$, and the highest by the USEPA [10] was $1900 \mu\text{g/l}$. For chromium, the highest TCLP concentration obtained by Eckert and Guo [35] was $986 \mu\text{g/l}$, the highest by the PCA [13] was $1540 \mu\text{g/l}$, and the highest by the USEPA [10] was $950 \mu\text{g/l}$.

The rates at which lead and chromium are dissolved have not been measured. An 18 h extraction time is required for the TCLP test. If we assume it takes 7 h to reach 95% saturation, then the dissolution coefficient (α) is 0.428 h^{-1} . A six-minute extraction time is required for the acid-digestion method. If we assume it takes four minutes to reach 95% saturation, the dissolution rate coefficient is 50 h^{-1} .

The molecular diffusion coefficient (D_m in Eq. (11)) for $1/2 \text{ Pb}^{2+}$ is $0.945 \times 10^{-9} \text{ m}^2/\text{s}$ [39]. The molecular diffusion coefficient for $1/3 \text{ Cr}^{3+}$ is $0.595 \times 10^{-9} \text{ m}^2/\text{s}$ [39]. The valency of chromium is more likely to be six rather than three under the high pH condition. The value of the molecular diffusion coefficient for $1/6 \text{ Cr}^{6+}$ has not been found in the literature.

The tortuosity (δ) for cement–mortar has not been measured. Van der Sloot and Kosson [40] measured the tortuosities of a number of different materials. The tortuosity of unconsolidated granular material was measured to be 2 to 5; that of light-weight concrete was 200 to 240; and that of concrete was 300 to 400. During measurements of tortuosity, sodium (an inert tracer) was used. Bentz and Garboczi [41] modeled the leaching of calcium hydroxide from cement paste and revealed the effects of leaching on pore space percolation and diffusivity. They found that for newly hydrated cement paste with $w:c$ ratio of 0.45, the effective diffusivity is 0.197% of the molecular diffusion coefficient. If 10% of the calcium hydroxide is leached out, the effective diffusivity is 1.7 times as much as that of newly hydrated paste. If 100% is leached, the effective diffusivity is 43 times as much as that of newly hydrated paste. From these results, using Eq. (11) and setting β equal to 1.0, the tortuosity of the newly hydrated cement paste is calculated to be 510. At 10% leaching, the tortuosity is reduced to 300. At 100% leaching, the tortuosity is reduced to 12. The tortuosity of cement–mortar is expected to fall between that of concrete and cement paste. It should be noted that a layer of calcium carbonate may form at the interface of cement–mortar lining and pipe water; this may lead to further physical retardation beyond that by the lining.

Concentrations of lead and chromium initially in the pipe water and those in incoming water at the upstream end ($C_{\text{pipe}0}$) vary widely, but they are expected to be below the drinking water standards ($15 \mu\text{g/l}$ for lead and $100 \mu\text{g/l}$ for chromium [42]).

5. Modeled results and assessment of potential increases of lead and chromium concentrations in static and flowing waters

5.1. Modeled lead and chromium concentrations in static pipe water

Eqs. (7), (8), (13) and (15) are solved simultaneously for the time variation of lead concentration in static pipe water, with input values of physical and chemical parameters. The following is the first set of physical and chemical parameters used in calculating the lead concentration in static pipe water (run number A1 in Table 2).

1. Inside diameter of the pipe (D) = 152 mm (6 in.);
2. Thickness of the lining (ΔR) = 6.4 mm (0.25 in.);
3. Water pressure head in the pipe (H_p) = 42.1 m (413 kPa, 60 psi);
4. Hydraulic conductivity (K) = 1.0×10^{-12} m/s;
5. Porosity of cement–mortar lining (η) = 0.3;
6. Initial moisture content inside the lining (θ_i) = 0.0;
7. Suction head inside the lining (H_s) = 0.703 m;
8. Available amount of lead in cement (C_a) = 28.1 mg/kg;
9. Sand-to-cement ratio (R_{sc}) = 2.0;
10. Water-to-cement ratio (R_{wc}) = 0.5;
11. Solid density of lining (ρ_s) = 2000 kg/m³;
12. Solubility (C_s , saturation concentration) of lead in the pore water = 70 $\mu\text{g/l}$;
13. Dissolution rate coefficient (α) = 0.428 h^{-1} ;
14. Molecular diffusion coefficient for lead (D_m) = $0.945 \times 10^{-9} \text{ m}^2/\text{s}$;
15. Tortuosity of cement–mortar lining (δ) = 200;
16. Initial concentration of lead in the pipe water ($C_{\text{pipe } 0}$) = 2.0 $\mu\text{g/l}$.

Values of porosity, hydraulic conductivity and tortuosity are expected to change with time and space because of the gradual dissolution of calcium hydroxide within the cement–mortar lining and the possible formation of calcium carbonate layer on the lining surface. The value of solubility is also expected to change with time and space because of variations of pH and chemical species. However, constant values of these parameters are used in this initial modeling.

The modeled time variation of the distance at which the wetting front has moved into the lining is shown in Fig. 3. The wetting front reaches the interface of the cast-iron pipe and the lining at 37.87 h (1.578 days).

The modeled time variation of lead concentration in the pipe is shown in Fig. 4. During the wetting process (time less than 1.578 days), the lead concentration increases more slowly than at the later stage, when the lining is fully wetted. This is because lead is advected into the lining at the same time it is diffused outward during the wetting process. After the lining is filled with water, the advection vanishes, and the lead concentration increases almost linearly with time. Lead concentration in the pipe

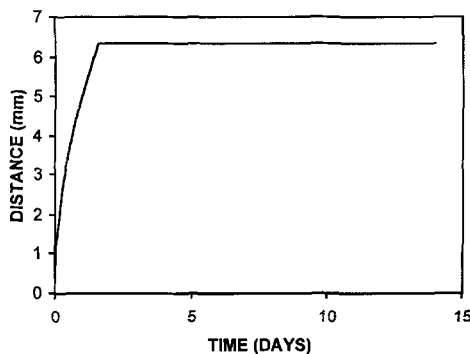


Fig. 3. Modeled time variation of distance to which a wetting front has moved into cement–mortar lining. The wetting front reaches the cast-iron pipe wall at 37.87 h (1.578 days).

Table 2
Effects of values of physical and chemical parameters on lead concentration in static pipe water

Run No.	D (mm)	ΔR (mm)	H_p (m)	K (m/s)	η	C_a (mg/kg)	C_s (ppb)	α (h^{-1})	δ	$C_{pipe,0}$ (ppb)	Time of full wetting (days)	C_{pipe} at the time of full wetting (ppb) ^c	C_{pipe} at seventh day (ppb) ^c	C_{pipe} at 14th day (ppb) ^c
A1 ^a	150	6.4	42.1	10^{-12}	0.3	28.1	70	0.43	200	2.0	1.58	3.02	9.03	16.02
A2				10^{-11}							0.16	2.02	9.60	16.50
A3				10^{-13}							> 14	N/A ^d	8.97	15.63
A4			14.3								4.58	6.11	8.73	15.75
A5							29				1.58	2.41	4.41	7.57
A6							1900				1.58	30.61	125.5	189.0
A7								50			1.58	11.20	26.84	36.22
A8								4.28			1.58	6.29	21.32	34.55
A9									100		1.58	3.64	11.95	21.18
A10									500		1.58	2.53	6.35	10.98
A11	200										1.58	2.75	7.23	12.59
A12	900										1.58	2.16	3.14	4.39
A13		3.2									1.58	3.47	9.25	15.99
A14						0.5					1.58	3.79	12.66	22.80
A15						0.2					1.58	2.76	6.65	11.35
A16						75.0					1.58	3.02	9.03	16.02
A17						0.25					1.58	2.35	3.34	4.34
A18										5.0	1.58	5.98	11.72	18.40
A19							9				1.58	2.11	2.72	3.44

^aValues of additional parameters not listed for run number A1 are: $\theta_i = 0.0$, $H_s = 0.70$ m, $R_{sc} = 2.0$, $R_{wc} = 0.5$, $\rho_s = 2000$ g/m³ and $D_m = 0.945 \times 10^{-9}$ m²/s.
^bUnspecified values of parameters are the same as those for run number A1.
^cThe action level for lead in drinking water is 15 ppb [42].
^dNot applicable.

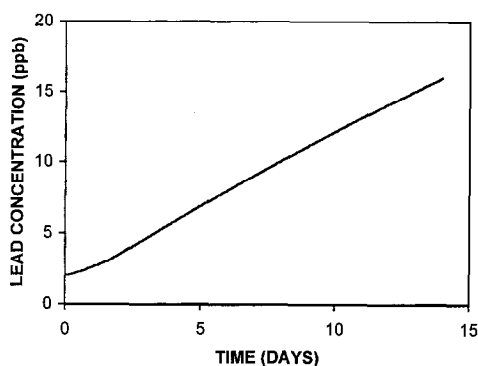


Fig. 4. Modeled time variation of lead concentration in static pipe water. The wetting front reaches the cast-iron pipe wall at 37.87 h (1.578 days).

increases from 3.02 ppb ($\mu\text{g}/\text{l}$) at the time of full wetting (1.578 days) to 9.03 ppb at the seventh day (Table 2), an increase of 1.11 ppb per day. The lead concentration increases from 9.03 ppb at the seventh day to 16.02 ppb at the 14th day (Table 2), an increase of 1.00 ppb per day. The rate at which lead leaches out to the pipe water decreases as the lead concentration in the pipe water increases, and the leaching of lead will cease when lead concentration in the pipe water increases to that of the pore water in the lining. The maximum possible concentration of lead in the pore water is the solubility (assumed to be 70 ppb in this first modeling case). The leaching of lead will also cease when the available amount of lead in the lining is exhausted. The amount of lead that remains in each computational segment of the lining is recalculated at each computational time step.

Fig. 5 shows the modeled time and spatial variations of lead concentration in the pore water inside the lining. During the wetting process (time less than 1.578 days), lead is dissolved, diffused, and advected in the wetted zone. The concentration is higher in the middle portion of the wetted zone than at both ends. Dissolved lead is diffused out to the

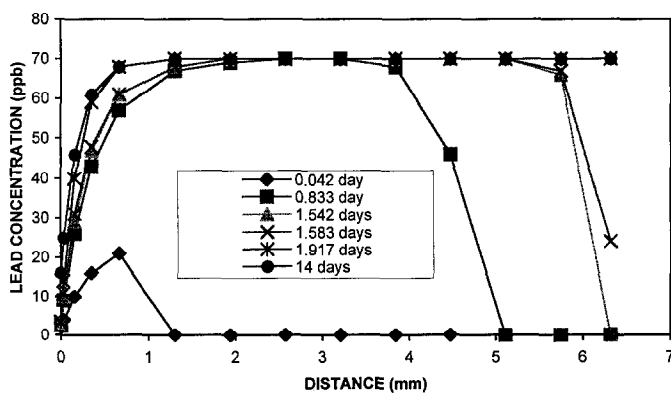


Fig. 5. Modeled spatial variations of lead concentration in pore water inside cement-mortar lining at six times instances before and after the full wetting. Refer to Fig. 4 for the corresponding lead concentration in pipe water.

pipe water at the interface of the pipe water and the lining. Dissolved lead is advected and diffused toward the dry zone at the wetting front. After the lining is fully wetted (time larger than 1.578 days), only diffusion occurs. Shortly after the full wetting, the concentration varies greatly in space near the pipe water–lining interface, but remains almost uniform in the interior zone. A leaching front is thus readily visible from this time on. As dissolved lead is diffused out to the pipe water, additional lead is dissolved from the solid lining matrix to replenish the pore water. Thus the concentration of lead in the pore water remains almost at the saturation concentration (solubility = 70 ppb in this first modeling case). The replenishment process will continue until available lead is exhausted. For this case, available lead has not been depleted at the end of the 14th day. At the later stage of the leaching process, lead concentration varies less in space near the pipe water–lining interface as the lead concentration of the pipe water increases, resulting in less lead diffusing out to the pipe water.

Values of other sets of physical and chemical parameters and corresponding modeled results are listed in Table 2. These results are compared with those of the first modeling case (run number A1) described above. In runs number A2 and A3, the hydraulic conductivity (K) is increased to 10^{-11} m/s and decreased to 10^{-13} m/s, respectively. With a larger value of hydraulic conductivity (10^{-11} m/s), the lining is filled with water faster, and concentrations at the seventh and 14th days are slightly larger. When the value of hydraulic conductivity is smaller (10^{-13} m/s), the lining is not yet fully wetted at the 14th day (the wetting front is at 5.99 mm, close to the lining thickness of 6.40 mm), and concentrations at the seventh and 14th days are slightly smaller. In run number A4, the pressure head of the pipe water (H_p) is decreased to 14.0 m (20 psi), and the lining is fully wetted at 109 h (4.58 days). Lead concentrations at the seventh and 14th days are smaller than those with higher pressure head. The larger the hydraulic conductivity and the water pressure are, the faster the water seeps into the lining (Eq. (7)). From these modeled results, it is observed that the faster the lining is filled with water, the faster lead leaches out.

If the solubility of lead in the pore water is larger, lead will leach out faster. In run number A5, when the solubility is decreased to 29 ppb (about one-half of that for run number A1), lead concentration in the pipe water is about one-half that of run number A1. In run number A6, when the solubility is increased to 1900 ppb (27 times that of run number A1), lead concentration of the pipe water increases greatly to about 12 times that of run number A1. The lead concentration does not increase proportionally because of depletion of lead in the lining near the pipe water–lining interface. In run number A19, when the solubility is reduced to 9 ppb, a minimal amount of lead is leached out within 14 days of water stagnation.

If lead dissolves faster, lead will leach out faster. In runs number A7 and A8, α is increased to 50 and 4.28, respectively. When α is increased from 0.428 h^{-1} to 4.28 h^{-1} (increased to 10 times that for run number A1), the lead concentration in the pipe water is doubled. When α is increased from 4.28 h^{-1} to 50 h^{-1} , the lead concentration in the pipe water increases much faster at the early stage of water stagnation, but does not increase much at the later stage of water stagnation because of the depletion of lead in the lining near the pipe water–lining interface and the solubility limit.

If the effective diffusion coefficient is increased, lead will leach out faster. In run number A9 in which tortuosity is decreased to 100 (thus from Eq. (11), the effective diffusion coefficient is two times that of run number A1), lead concentration in the pipe water is 1.33 times that of run number A1. In run number A10, where tortuosity is increased to 500 (thus the effective diffusion coefficient is 40% of that for run number A1), lead concentration in the pipe water is 68.5% of that for run number A1.

If the pipe diameter is larger, the concentration of lead in the pipe water is smaller because of dilution. This is demonstrated by the results from runs number A11 and A12. The thickness of the lining has almost no effect on the rate at which lead leaches out before the depletion. This is demonstrated by the results from run number 13. The larger the porosity of the lining, the faster lead leaches out because more lead is dissolved in the water. This is demonstrated by the results from runs number A14 and A15.

In run number A17, because only a smaller amount of lead is available in cement (0.25 mg/kg), lead is depleted near the pipe water–lining interface during the early stage of leaching. Thus, lead leaches out more slowly, and the lead concentration in the pipe water increases more slowly. In run number A16, a larger amount of lead (75 mg/kg) is available than in run number A1; however, the same amounts of lead leaches out within 14 days of water stagnation as in run number A1. This is because, in both cases, lead has not been depleted during the modeling period (14 days).

If the initial concentration of lead in the pipe water is larger, lead will leach out more slowly because of the smaller concentration gradient at the pipe water–lining interface, resulting in a slower diffusion process. This is demonstrated by the results of run number A18.

The sensitivities of the modeled results to two numerical parameters were tested. In all of the above modelings, the thickness of the lining is divided uniformly into 100 computational elements, and the time interval of modeling is 0.01 h. When the thickness is divided into 200 computational elements, rather than 100 for run number A1 in Table 2, lead concentrations at the time of full wetting, on the seventh day and on the 14th day are 2.99 ppb, 9.05 ppb and 16.10 ppb, respectively. They are only slightly different from those of run number A1 (less than 1% difference). This demonstrates that the modeled concentration is not sensitive to the number of computational elements if the number is larger than 100. When the computational time interval is changed from 0.01 h to 0.005 h for run number A1 in Table 2, no change in modeled lead concentration is noticed. Therefore, a sufficiently large number of computational elements and a sufficiently small computational time interval are used in all modelings described above.

The modeled chromium concentrations in the static pipe water with various values of physical and chemical parameters are listed in Table 3. Changes in modeled chromium concentrations in response to changes in values of physical and chemical parameters are similar to those for lead in Table 2.

5.2. Modeled lead and chromium concentrations in flowing pipe water

Eqs. (7), (8), (13) and (18) are solved simultaneously for time and spatial variations of lead and chromium concentrations in the flowing pipe water, with input values of physical and chemical parameters. All physical and chemical parameters that were used for the modelings of the static pipe water are required for the flowing water modelings.

Table 3

Effects of physical and chemical parameters on chromium concentration in static pipe water

Run number	C_a (mg/kg)	C_s (ppb)	δ	$C_{\text{pipe } 0}$ (ppb)	Time of full wetting (days)	C_{pipe} at the time of full wetting (ppb) ^c	C_{pipe} at seventh day (ppb) ^c	C_{pipe} at 14th day (ppb) ^c
B1 ^a	76	540	200	15.0	1.58	20.71	57.75	101.82
B2	422	1540			1.58	31.60	139.17	267.09
B3	422	6160			1.58	81.89	515.36	985.37
B4	422	7800			1.58	99.74	648.90	1163.87
B5	28.0	986		7.3	1.58	17.98	77.11	116.54
B6	^b		500		1.58	17.89	40.08	69.63
B7			1000		1.58	17.32	37.03	61.45
B8	25	70			1.58	15.6	19.48	24.10

^aValues of additional parameters not listed for run number B1 are: $D = 150$ mm, $\Delta R = 6.4$ mm, $H_p = 42.1$ m, $K = 1.0 \times 10^{-12}$ m/s, $\theta_i = 0.0$, $H_s = 0.70$ m, $R_{sc} = 2.0$, $R_{wc} = 0.5$, $\rho_s = 2000$ kg/m³, $\alpha = 0.428$ h⁻¹ and $D_m = 0.595 \times 10^{-9}$ m²/s.

^bUnspecified values of parameters are the same as those for run number A1.

^cThe maximum contaminant level for chromium in drinking water is 100 ppb [42].

Additional parameters for the modelings of flowing water are flow velocity (U_{pipe}) and pipe length.

In the first modeling case for lead concentration in flowing water (run number C1 in Table 4), the flow velocity is 0.6 m/s, the pipe length is 48.3 km (30 miles), and the rest of the parameters have the same values as those for run number A1 in the static water case (Table 2). The modeled results, along with the values of those adjusted parameters, are listed in Table 4. The modeled time variations of lead concentrations at 4.83 km and 48.3 km from the upstream end of the pipe are shown in Fig. 6. The lead concentration in flowing water is much smaller than that in static pipe water because of dilution. After a short while, the lead concentration in flowing water does not increase with time since the flowing water carries lead downstream as fast as lead is leached out from the lining (i.e., leached lead is not accumulated in the pipe water). The modeled

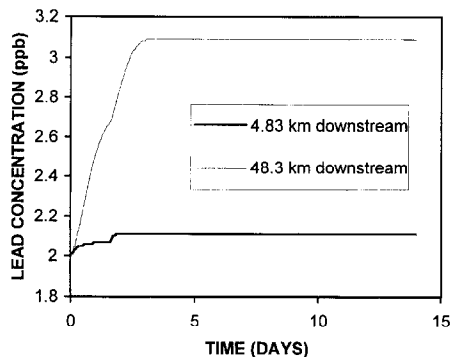


Fig. 6. Modeled time variations of lead concentration in flowing pipe water at two locations.

Table 4
Effects of physical and chemical parameters on lead concentration in flowing pipe water

Run No.	U_{pipe} (m/s)	C_s (ppb)	C_a (mg/kg)	C_{pipe} at 4.83 km downstream at the time of full wetting, 1.58 days (ppb) ^b	C_{pipe} at 4.83 km downstream at seventh day (ppb) ^b	C_{pipe} at 4.83 km downstream at 14th day (ppb) ^b	C_{pipe} at 4.83 km downstream at the time of full wetting, 1.58 days (ppb) ^b	C_{pipe} at 4.83 km downstream at seventh day (ppb) ^b	C_{pipe} at 4.83 km downstream at 14th day (ppb) ^b
C1 ^a	0.6	70	28.1	2.07	2.11	2.11	2.68	3.09	3.09
C2	1.25	70	28.1	2.04	2.05	2.05	2.35	2.53	2.53
C3	0.6	1900	28.1	4.07	3.07	2.76	20.92	13.45	9.93
C4	1.25	1900	28.1	3.01	2.52	2.36	11.70	7.26	5.73
C5	0.6	1900	530	4.07	5.07	5.07	20.93	32.45	32.45

^aAdditional parameters have the same values as those for run number A1 in Table 2.

^bThe action level for lead in drinking water is 15 ppb [42].

Table 5
Effects of physical and chemical parameters on chromium concentration in flowing pipe water

Run No.	U_{pipe} (m/s)	C_s (ppb)	C_a (mg/kg)	C_{pipe} at 4.83 km downstream at the time of full wetting, 1.58 days (ppb) ^b	C_{pipe} at 4.83 km downstream at seventh day (ppb) ^b	C_{pipe} at 4.83 km downstream at 14th day (ppb) ^b	C_{pipe} at 4.83 km downstream at the time of full wetting, 1.58 days (ppb) ^b	C_{pipe} at 4.83 km downstream at seventh day (ppb) ^b	C_{pipe} at 4.83 km downstream at 14th day (ppb) ^b
fsD1 ^a	0.6	1540	422	16.22	16.94	16.94	26.06	34.30	34.30
fsD2	1.25	1540	422	15.59	15.93	15.93	20.69	24.31	24.31
fsD3	0.6	6160	422	19.91	22.82	20.62	59.56	92.79	70.97
fsD4	1.25	6160	422	17.39	18.76	17.70	37.94	52.52	41.96
fsD5	0.6	7800	422	21.22	24.91	20.56	71.45	113.56	70.39

^aAdditional parameters have the same values as those for run number B1 in Table 3.

^bThe maximum contaminant level for chromium is 100 ppb [42].

constant lead concentrations at 4.83 km and 48.3 km are 2.11 ppb and 3.09 ppb, respectively. Lead concentration downstream is higher than that at the upstream section because additional lead is leached out along the pipe.

In the second run (Table 4), the flow velocity is increased to 1.25 m/s, and the constant lead concentrations of 4.83 km and 48.3 km downstream are decreased to 2.05 ppb and 2.53 ppb, respectively, which are smaller than those for run number C1 because of a higher dilution ratio. In the third and fourth runs (Table 4), the solubility is increased to 1900 ppb; this yields maximum lead concentrations in the pipe water of 20.92 ppb and 11.70 ppb, respectively. These concentrations are much higher than those for run number C1. However, concentration decreases with time because of the depletion of lead near the pipe water–lining interface. When the lead availability is increased to a much larger value in run number C5, lead in the lining is not depleted, and lead concentration remains constant within the computational time period.

The modeled chromium concentrations in flowing pipe water with various values of physical and chemical parameters are listed in Table 5. The total pipe length is divided into 10 computational segments in the modelings.

5.3. Assessment of potential increase in lead concentration from using cement–mortar-lined pipes

All values used in run number A1 (Table 2) are possible values in the field. However, it must be noted that the diameter of 150 mm is the smallest possible in the field, and the tortuosity (200) is also likely to be the lowest possible in the field.

From the sensitivity studies performed (Table 2), it is observed that the rate of lead leaching from the lining is most sensitive to the solubility of lead in the pore water inside the cement–mortar lining if the lead is not depleted. The TCLP concentration was argued in Section 4.3 to be one estimate of lead solubility in pore water. When the average TCLP concentration (9 ppb) measured by the PCA [13] is used as the solubility (run number A29), the lead concentration will increase only by 0.1 ppb per day of water stagnation. When the maximum TCLP concentration (29 ppb) measured by the PCA [13] is used as the solubility (run number A5), the lead concentration in the pipe water will increase by about 0.4 ppb per day of water stagnation. These rates of increase of lead concentration are unlikely to cause the exceedance of the action level (15 ppb) in the pipe water even with a long water stagnation time.

When the solubility is increased to 70 ppb (run number A1), lead concentration in the pipe water will increase by about 1.0 ppb per day of water stagnation. Solubility was estimated in Section 4.3 to be from one to four or more times the TCLP concentration. 70 ppb is about two and one-half times that of the maximum TCLP concentration measured by the PCA [13] and is also the solubility of lead in drinking water at a high pH [37]. With the lead concentration increase of 1.0 ppb per day of water stagnation, exceedance of the action level is possible only if water is stagnant for a long period of time (e.g., two weeks at the dead ends of the pipes). If water is flowing at the normal velocity, the maximum increase of lead concentration is only about 1.0 ppb (runs number C1 and C2 in Table 4) even with the contribution of lead from the lining of a 48.3-km pipe.

If the maximum TCLP concentration (1900 ppb) measured by the USEPA [10] is used as the solubility (run number A6), the lead concentration in the pipe water will increase by 17.5 ppb per day of water stagnation. With such a high solubility value, even the leached lead is diluted by the flowing water, and the action level will still be exceeded (runs number C3 and C5). However, the USEPA's high TCLP concentration remains to be confirmed, and is unlikely to be representative of cement samples.

Colucci et al. [15] conducted tests on the amounts of toxic metals that may leach out from cement–mortar cubes following the procedure proposed by the National Sanitation Foundation (NSF) [43]. The surface area-to-volume ratio of the cube was equivalent to that of an 88-mm diameter pipe. The test cube was submersed (conditioned) in water for 14 days, and the water was changed 10 times during that period. After the conditioning, the individual cube was submersed in extraction water for 72 h, and concentrations of toxic metals in this latter extraction water were measured. The maximum lead concentration in the extraction water was less than the detection limit ($1.0 \mu\text{g/l}$). Neither acid-soluble nor TCLP concentration of lead in the cement used was reported.

Kanare and West [16], also following the NSF test procedure [43], tested leachability of toxic metals from concrete cylinders. The surface area-to-volume ratio of the test cylinder was equivalent to that of a 910-mm diameter pipe. The maximum lead concentration of the extraction water was measured to be $2.1 \mu\text{g/l}$. The cement used in making this particular cylinder had an acid-soluble lead content of 8.11 mg/kg and TCLP lead concentration of a value below the detection limit ($5 \mu\text{g/l}$). For another cylinder made of cement with acid-soluble lead content of $< 5 \text{ mg/kg}$ and TCLP concentration of $7 \mu\text{g/l}$, the lead concentration in the extraction water was measured to be $1.2 \mu\text{g/l}$. One control water sample had a lead concentration of $1.0 \mu\text{g/l}$; another had a lead concentration of $5.8 \mu\text{g/l}$, which is higher than those of any extraction waters. It appears that lead concentrations in extraction water increased by 0 to $1.1 \mu\text{g/l}$ after one day of extraction.

Germaneau et al. [17], following a test procedure similar to the NSF procedure [43], conducted the leachability of heavy metals from cement–mortar bars in contact with drinking water. The bar had a volume of water to contact surface ratio of 5 cm, a value typical of pipes having a 100 mm internal diameter. The bar was immersed in tap water for 24 h, in tap water with 100 mg/l chlorine for 24 h, then in tap water for another 24 h, and finally in Evian water for two separate 24 h periods. Among bars made of ordinary portland cement (OPC), one bar had lead concentrations in the five immersion (extraction) fluids of 3, 12, < 1 , 2 and $< 1 \mu\text{g/l}$, respectively. It is noted that the lead concentration ($12 \mu\text{g/l}$) in the chlorinated water was much higher than those in tap and Evian waters. The OPC used in making this cylinder had an acid-soluble lead content of $< 10 \text{ mg/kg}$. For another bar made of OPC with an acid-soluble lead content of 8.6 mg/kg , lead concentrations of the five immersion waters were 2, 2, 1, < 1 and $< 1 \mu\text{g/l}$, respectively. Lead concentration of the chlorinated water was not much higher than that of other waters for this bar. Lead concentrations originally in the extraction fluids were not reported.

The leaching environment for the cement–mortar linings inside drinking water pipes is not exactly the same as that for cement–mortar cubes and cylinders. For example, the cement–mortar lining is subject to pressure. This may result in a slightly larger amount

of lead being leached out based on the parametric studies described in Section 5.1, if all other factors remain the same. Nevertheless, the test results described above are used for comparison with the modeled results.

In the tests conducted by Kanare and West [16], $D = 910$ mm and $C_a = 7.01$ mg/kg. If the tortuosity (δ) is assumed to be 200 for the newly hydrated concrete, the solubility (C_s) is assumed to be the same as the TCLP concentration (6.5 ppb), and the rest of the parameters are assumed to be the same as those in run number A1 (Table 2), the solution to the governing equations yields an increase of 0.013 ppb lead concentration per day of water stagnation (extraction). If the solubility is assumed to be four times the TCLP concentration (26 ppb), the modeled lead concentration increase rate is 0.064 ppb per day of water stagnation. These modeled increases are in the range of those measured (0 to 1.1 ppb per day).

In the tests conducted by Germaneau et al. [17], $D = 100$ mm and $C_a = 8.6$ mg/kg. If the TCLP concentration is assumed to be 7.1 ppb based on the mean ratio of TCLP concentration and acid-soluble content in cement [13], the solubility (C_s) is assumed to be the same as the TCLP concentration, and the rest of the parameters are assumed to be the same as those in run number A1 (Table 2), the solution to the governing equations yields an increase of 0.12 ppb lead concentration per day of water stagnation (extraction). If the solubility is assumed to be four times the TCLP concentration (28.4 ppb), the modeled lead concentration increase rate is 0.62 ppb per day of water stagnation. These modeled increase rates are in the same order as those tested (< 1 to 3 ppb per day, excluding that for the chlorinated water).

The pilot laboratory results of Guo et al. [18] showed that detectable amounts of lead leached out from the cement–mortar lining. In one test pipe, the lead concentration was measured to increase from 3.08 to 13.52 ppb within three days of water stagnation. However, this large increase rate (3.48 ppb per day) was not reproduced by tests on the other two pipes (0.01 and 0.48 ppb per day, respectively). The laboratory test conditions were similar to that of run number A1 in Table 2. The modeled increase in lead concentration (1 ppb per day) is in the same order as those tested (0.01 to 3.48 ppb per day).

Based on the modeled results (run number A1 in Table 2), cement with lead solubility larger than 70 ppb at a pH equal to that of drinking water is not recommended for in-place lining of drinking water pipes. This recommendation on the maximum allowable solubility (70 ppb) is based on the projection that the action level of lead in drinking water (15 ppb) will be exceeded after the initial 14 days of water stagnation. At the same lead solubility (70 ppb), if the pipe water is flowing at the normal velocity (0.6 m/s), lead concentration will increase only by 1.09 ppb (run number C1, Table 4), which is far below the action level (15 ppb). However, the projected increase of lead concentration in the flowing pipe water (1.09 ppb) is not much lower than the maximum allowable level (MAL) (1.5 ppb, 10% of the action level) recommended by the NSF [43].

When the lead solubility is not available, the TCLP concentration could be used in selecting cement. If the solubility is assumed to be from one to four times the TCLP concentrations, cement with TCLP concentrations of lead less than 18 ppb is very likely to be safe, but cement with TCLP concentration larger than 70 ppb is not recommended for lining drinking water pipes.

The modeled results based on measured TCLP concentrations, along with the limited laboratory data available, indicate that the possibility is small that the action level of lead will be exceeded by using in-place cement–mortar-lined pipes at the level of HWF-burning prior to 1992. Among the PCA's [13] 97 cement samples, 3 had TCLP concentrations larger than 18 ppb (the very safe concentration), but all had TCLP concentrations less than 70 ppb (the maximum recommended concentration).

5.4. Assessment of potential increase in chromium concentration from using cement–mortar-lined pipes

When the PCA's [13] measured mean TCLP concentration (540 $\mu\text{g}/\text{l}$) is used as the solubility and the mean acid-soluble content (76 mg/kg) is used as the availability (run number B1 in Table 3), chromium concentration increases by 6.8 ppb per day of water stagnation. With this rate of chromium concentration increase, the maximum containment level (MCL) for chromium (100 ppb) is likely to be exceeded if water is stagnant for a long period of time (e.g., 14 days). When the PCA's [13] maximum TCLP concentration (1540 ppb) is used as the solubility and the maximum acid-soluble content (422 mg/kg) is used as the availability, chromium concentration increases by 19.8 ppb per day of water stagnation. With this large rate of chromium concentration increase, the chromium concentration is likely to be exceeded in static pipe water in 5 days. If water is flowing at normal velocities from 0.6 to 1.25 m/s, the chromium concentrations in the pipe water will only be 24.3 to 34.3 ppb, which is lower than the MCL for chromium (runs number D1 and D2 in Table 5). However, if the solubility is four times the maximum measured TCLP concentration, the chromium concentration will increase by 80 ppb per day of water stagnation (run number B3 in Table 3), and the maximum concentration will rise to 92.8 ppb in the flowing pipe water (runs number D3 and D4 in Table 5).

The maximum chromium concentration in extraction water measured by Colucci et al. [15] was 3 ppb. In Kanare and West's [16] tests for a concrete cylinder made of cement with acid-soluble chromium content of 99.3 mg/kg and TCLP concentration of 1050 ppb, the chromium concentration in the extraction water was 0.3 ppb, only 0.1 ppb larger than that of the controlled sample. For another concrete cylinder made of cement with an acid-soluble chromium content of 167 mg/kg and a TCLP concentration of 1220 ppb, the chromium concentration in the extraction water was also 0.3 ppb. These test results are much less than those modeled (on the order of 10 to 15 ppb per day of extraction). In Germaneau et al.'s [17] tests, for one mortar bar made of OPC with an acid-soluble chromium content of 125 mg/kg, the chromium concentration of five extraction fluids were 12 ppb, 1 ppb, < 1 ppb, < 1 ppb and < 1 ppb, respectively. For another bar made of OPC with an acid-soluble chromium content of 100 mg/kg, chromium concentrations in five extraction fluids were 21 ppb, 6 ppb, 1 ppb, 2 ppb and < 1 ppb, respectively. These test results are in the area of that modeled (about 10 ppb).

Results of laboratory tests by Guo et al. [18] appear to support the modeled results. For one test pipe, chromium concentration increased from 0.49 ppb to 8.58 ppb within the first day of water stagnation. This represented a concentration increase of 8.09 ppb

per day of water stagnation. The cement used in lining the pipe had an acid-soluble chromium content of 28 mg/kg and a TCLP concentration of 986 ppb. The pipe had a diameter of 150 mm (6 in.), and the thickness of the lining was 6.4 mm (1/4 in.). A modeling was carried out based on these data (run number B5 in Table 3). The modeled chromium concentration increased by 10.9 ppb per day of water stagnation, which is similar to that measured. For the other two test pipes, chromium concentrations increased by 0.82 ppb per day and 9.66 ppb per day, respectively. Chromium concentrations in all three test pipes did not increase much after one day of water stagnation. This may have been caused by reprecipitation of chromium, since the pH in the pipe water also increased with time.

Based on the modeled results (run number B1 in Table 3), cement with chromium solubility larger than 540 ppb at a pH equal to that of drinking water is not recommended for in-place lining of drinking water pipes. This recommendation on the maximum allowable solubility (540 ppb) is based on the projection that the MCL in drinking water (100 ppb) will be exceeded after the initial 14 days of water stagnation. The projection is based on the assumption that chromium concentration in the pipe water will always increase with stagnation time. However, as Guo et al.'s [18] pilot laboratory results indicated, chromium concentration did not increase much with stagnation time after the first day. If only one day stagnation time rather than two weeks is used, cement with a chromium solubility higher than 7800 ppb (run number B4 in Table 3) is not recommended for use in lining the pipe.

When the chromium solubility is not available, the TCLP concentration could be used in selecting cement. If the solubility is assumed to be from one to four times the TCLP concentration, cement with TCLP concentrations of chromium less than 135 ppb is very likely to be safe, but cement with TCLP concentrations larger than 540 ppb is not recommended for lining drinking water pipes. If the possible reprecipitation in static water or the flowing condition is considered, cement with a TCLP concentration larger than 7800 ppb is not recommended.

The modeled results based on measured TCLP concentrations, along with the limited laboratory data available, indicate that there is a moderate possibility that the MCL for chromium will be exceeded by using in-place cement–mortar-lined pipes at the HWF-burning level prior to 1992. Among the PCA's [13] 97 cement samples, only five samples had mean TCLP concentrations lower than 135 ppb (the very safe concentration), and 37 samples had a TCLP concentration larger than 540 ppb (the maximum recommended nonreprecipitation controlled concentration). However, none of the samples had TCLP concentrations larger than 7800 ppb (the maximum recommended reprecipitation controlled concentration).

It should also be pointed out that the use of currently available cement could cause the MAL recommended by the NSF [43] to be exceeded. If the maximum TCLP chromium concentration (1540 ppb) measured by PCA [13] is used as the solubility and if the pipe water is flowing at the normal velocity (0.6 m/s), chromium concentration will increase by 9.3 ppb (run number D1, Table 5), which is very close to the MAL (10 ppb, 10% of the MCL) recommended by the NSF [43].

6. Implications for the practice of using HWF in cement kilns

Eckert and Guo [35] found a significant correlation between the TCLP concentration and the acid-soluble content of lead in cement among the limited 16 samples analyzed. The relevant statistical parameters [44] are: correlation coefficient (r_{xy}) = 0.89, $N = 6$ (only six samples had detectable lead TCLP concentrations), and $[(N - 3)^{0.5}/2] \ln[(1 + r_{xy})/(1 - r_{xy})] = 2.46 > 1.96$. Based on Eckert and Guo's [35] data, if the intercept (b) is forced to be zero, the following relation between the TCLP concentration and the acid-soluble content of lead in cement can be obtained by the least squares fit:

$$\begin{aligned} \text{Lead TCLP concentration } (\mu\text{g l}^{-1}) \\ = 0.630 \text{ Lead acid-soluble content } (\text{mg kg}^{-1}). \end{aligned} \quad (20)$$

If the intercept is not forced to be zero, the fitted b value is 2.84 and the fitted slope (a) value is 0.50. However, the PCA's [13] study indicated an insignificant correlation between the TCLP concentration and the acid-soluble content of lead in cement among the 97 samples analyzed [correlation coefficient (r_{xy}) = 0.052, $N = 36$, $[(N - 3)^{0.5}/2] \ln[(1 + r_{xy})/(1 - r_{xy})] = 0.30 < 1.96$]. The reason for this discrepancy is unknown. The correlation relation (Eq. (20)) obtained from Eckert and Guo's [35] data is used in this initial assessment. From this relation, the very safe lead TCLP concentration for cement (18 ppb) corresponds to the acid-soluble content of 29 mg/kg, and the maximum recommended lead TCLP concentration for cement (70 ppb) corresponds to the acid-soluble content of 111 mg/kg.

For the kiln studied by Guo and Eckert [12], at the maximum allowable lead content in HWF fed to the kiln, the acid-soluble content of clinker is only 12.3 mg/kg, while CKD had an acid-soluble content of 2740 mg/kg. During the trial burn, 40% of CKD was recirculated back to the kiln. Based on Eq. (20), the lead TCLP concentration corresponding to an acid-soluble content of 12.3 mg/kg is only 7.7 ppb. Use of cement with such a low lead TCLP concentration is unlikely to cause the exceedance of the action level of lead in drinking water, as demonstrated in the models in Sections 5.1 and 5.2. For a daily composite sample of wasted CKD with an acid-soluble content of 2030 mg/kg, the TCLP concentration was measured to be 1.3 ppm [45]. After adjusting the acid-soluble content to 2740 mg/kg, the TCLP concentration would be 1.8 ppm, which is lower than the RCRA limit for lead (5.0 ppm). The wasted CKD would not be classified as hazardous waste based on the TCLP leachable amount of lead.

However, if 100% of the CKD were recirculated back to the studied kiln, the acid-soluble content of clinker would be 430 mg/kg (calculated from the data given above). Using Eq. (20), the corresponding TCLP concentration would be 271 ppb. This TCLP concentration is much higher than the maximum recommended TCLP concentration (70 ppb). Therefore, at the maximum allowable lead content in HWF feed, 100% CKD recirculation is not recommended.

Both the PCA [13] and Eckert and Guo [35] found a significant correlation between the acid-soluble content and the TCLP concentration of chromium in cement. The correlation coefficient (r_{xy}) was found to be 0.63 among the PCA's [13] 97 samples [$N = 92$, $[(N - 3)^{0.5}/2] \ln[(1 + r_{xy})/(1 - r_{xy})] = 7.15 > 1.96$]. The correlation coeffi-

cient (r_{xy}) was found to be 0.73 among Eckert and Guo's [35] 16 samples [$N = 16$, $[(N - 3)^{0.5}/2] \ln[(1 + r_{xy})/(1 - r_{xy})] = 3.35 > 1.96$]. Based on Eckert and Guo's [35] data and if the intercept is forced to be zero, the relation between the TCLP concentration and the acid-soluble content can be obtained by the least squares fit as:

$$\begin{aligned} &\text{Chromium TCLP concentration } (\mu\text{g l}^{-1}) \\ &= 17.1 \text{ Chromium acid-soluble content } (\text{mg kg}^{-1}). \end{aligned} \quad (21)$$

If the intercept is not forced to be zero, the fitted intercept (b) value is -141.85 and the fitted slope (a) is 26.13 . From the correlation relation (Eq. (21)), the very safe chromium TCLP concentration for cement (135 ppb) corresponds to the acid-soluble content of 9 mg/kg, the recommended maximum nonprecipitation controlled TCLP concentration for cement (540 ppb) corresponds to the acid-soluble content of 32 mg/kg, and the recommended maximum reprecipitation controlled TCLP concentration for cement (7800 ppb) corresponds to the acid-soluble content of 456 mg/kg.

For the kiln studied by Guo and Eckert [12], at the maximum allowable chromium content in HWF fed to the kiln, the acid-soluble content of clinker was 483 mg/kg, while CKD had the acid-soluble content of 409 mg/kg. Based on Eq. (20), the TCLP concentration corresponding to the acid-soluble content of 483 mg/kg is 8260 ppb, which is higher than the highest recommended TCLP concentration (7800 ppb). Such a cement is not recommended to be used for the lining of drinking water pipes. For a daily composite sample of wasted CKD with an acid-soluble content of 386 mg/kg, the TCLP concentration was measured to be 5100 ppb [45]. After the adjustment of the acid-soluble content to 483 mg/kg, the TCLP concentration would be 6400 ppb, which is significantly higher than the RCRA limit for chromium (5000 ppb). The wasted CKD could have been classified as hazardous waste.

If 100% of CKD were recirculated back to the studied kiln, the acid-soluble content of chromium in the clinker would be reduced to 464 mg/kg (calculated from the data given above). Using Eq. (20), the corresponding TCLP concentration would be 7920 ppb. This TCLP concentration is still higher than the highest recommendation (7800 ppb). Therefore, for this particular kiln studied, it is recommended that the maximum allowable chromium fed to the kiln be lowered. It can be achieved by either reducing the chromium content in HWF or by reducing the overall amount of HWF feed.

7. Conclusions

A mathematical leaching model was developed as a tool for assessing the potential increases of lead and chromium in drinking water from using cement–mortar-lined pipes. The initial wetting process, dissolution, diffusion and advection of metals in the lining, accumulation of metals in the static pipe water and dilution of metals by the flowing water were considered. Values for physical and chemical parameters in the model were obtained from literature or were estimated.

Based on the modeled results, if the solubility of lead (at a pH equal to that of drinking water) in cement is larger than 70 ppb, the use of cement–mortar-lined pipes

could cause the action level of lead in drinking water (15 ppb as established by the USEPA) to be exceeded. At the level of HWF burning in cement kilns prior to 1992, the action level for lead in drinking water is unlikely to be exceeded by using cement–mortar-lined pipes.

Also based on the modeled results, if the solubility of chromium (at a pH equal to that of drinking water) in cement is larger than 540 ppb, the use of cement–mortar-lined pipes could cause the MCL of chromium in drinking water to be exceeded. At the level of HWF burning in cement kilns prior to 1992, the MCL of chromium (100 ppb as established by the USEPA) is likely to be exceeded by using cement–mortar-lined pipes if water is stagnant for two weeks and if no reprecipitation of chromium occurs. At this level (1992 level) of HWF burning and if the pipe water is flowing at the normal velocity, the MCL of chromium (100 ppb) as established by the USEPA is unlikely to be exceeded, but the MAL (10 ppb) as established by the NSF is likely to be exceeded.

As a precaution at this time, cement with a TCLP concentration of lead larger than 70 ppb or an acid-soluble content of lead larger than 111 mg/kg is not recommended to be used for the lining of drinking water pipes; cement with a chromium TCLP concentration larger than 540 ppb or an acid-soluble content larger than 32 mg/kg is also not recommended. For one cement kiln studied, when HWF has the maximum allowable lead content, it is recommended that CKD not be fully recirculated. It is also recommended that the maximum allowable chromium content of HWF feed be lowered. A number of assumptions were made in developing these initial recommendations. These assumptions remain to be justified using data from future comprehensive laboratory and/or field tests. The leaching model developed and the initial assessment made in this paper provides guidance for the design of these future tests.

8. Nomenclature

C_a	Availability of metal based on the mass of cement (mg/kg)
$C_{a \text{ mortar}}$	Amount of metal in cement–mortar lining available for leaching on the mass basis (mg/kg)
C_{pipe}	Concentration of metal in the pipe water ($\mu\text{g/l}$)
C_{pore}	Metal concentration of the pore water ($\mu\text{g/l}$)
C_s	Solubility (saturation concentration) of metal compound ($\mu\text{g/l}$)
D	Inside diameter of the pipe (mm or in.)
D_e	Effective diffusion coefficient (m^2/s)
$\frac{dH}{dr}$	Gradient of pressure head (H) in the radial (r) direction
F	Amount of water per unit length of the pipe that has seeped into the lining ($1/\text{m}$)
H_p	Pressure head of the pipe water (m)
H_s	Suction head of cement–mortar lining before the wetting (m)
J	Net amount of metal which is leaching out from the lining surface per unit time per unit length of the pipe ($\mu\text{g}/\text{m}/\text{s}$)
K	Hydraulic conductivity of the cement–mortar lining (m/s)
k	Permeability of the cement–mortar lining (m^2)

S	Metal content of the cement–mortar on the mass basis (mg/kg)
R_{sc}	Sand-to-cement ratio on the mass basis
R_{wc}	Water-to-cement ratio on the mass basis
r	Distance in the radial direction (m)
U_{pipe}	Water flow velocity (m/s)
U_s	‘Apparent’ water seepage velocity in the cement–mortar lining (m/s)
x	Distance along the pipe (m)
T_a	Total time period during which the available amount of metal is depleted (s)
T_s	Total time period the water takes to fill the pore space in the cement–mortar lining (s)
t	Time (s)
α	Dissolution rate coefficient (h^{-1})
γ	Specific weight of water (kN/m^3)
ΔR_w	Distance at which the wetting front has moved into the lining (m)
δ	Tortuosity
η	Porosity of the cement–mortar lining
θ_i	Initial moisture content of the lining
μ	Viscosity of water ($\text{Pa} \cdot \text{s}$)
ρ_s	Density of cement–mortar lining based on the volume of solid (kg/m^3)

Acknowledgements

P. Toomuluri and J.O. Eckert, Jr. assisted in literature research and review. Informative discussions with P.N. Balaguru and S.J. Medlar are acknowledged. D.S. Kosson is thanked for introducing his related research work. Preliminary, unpublished work by W. Ziegler and H.K. Roffman stimulated the author’s interest in this research topic. The Missouri Department of Natural Resources provided a full copy of the CoC report. The research was supported financially in part by the Association for Responsible Thermal Treatment (ARTT), Washington, D.C. Although the study was sponsored by ARTT, it does not necessarily reflect the views of ARTT and no official endorsement should be inferred.

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