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Permeation and Leaching

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Background and Disclaimer

The USEPA is revising the Total Coliform Rule (TCR) and is considering new possible distribution system requirements as part of these revisions. As part of this process, the USEPA is publishing a series of issue papers to present available information on topics relevant to possible TCR revisions. This paper was developed as part of that effort.

The objectives of the issue papers are to review the available data, information and research regarding the potential public health risks associated with the distribution system issues, and where relevant identify areas in which additional research may be warranted. The issue papers will serve as background material for EPA, expert and stakeholder discussions. The papers only present available information and do not represent Agency policy. Some of the papers were prepared by parties outside of EPA; EPA does not endorse those papers, but is providing them for information and review.

Additional Information

The paper is available at the TCR web site at:

   http://www.epa.gov/safewater/disinfection/tcr/regulation_revisions.html

Questions or comments regarding this paper may be directed to **TCR@epa.gov.**

Permeation & Leaching

1.0   General Description of Topics

Distribution system infrastructure and appurtenances, including piping, linings, fixtures, and solders, can react with the water they supply as well as the external environment. These interactions can result in degradation of the distributed water. Permeation of plastic pipes and leaching from linings and metal appurtenances are known pathways for water quality degradation.

Permeation of piping materials and non-metallic joints can be defined as the passage of contaminants external to the pipe, through porous, non-metallic materials, into the drinking water. The problem of permeation is generally limited to plastic, non-metallic materials.

Leaching can be defined as "the dissolution of metals, solids, and chemicals into drinking water" (Symons et al, 2000). Leaching can result in elevated levels of metals, organic contaminants, or asbestos in water consumed at the tap. Health effects and mitigation techniques related to leaching of lead and copper from lead service lines or household plumbing materials are addressed in the Lead and Copper Rule (USEPA, 1991). Health effects associated leaching of asbestos fibers from asbestos-cement piping is currently addressed under the Phase II National Primary Drinking Water Regulations (USEPA, 1991). Thus, this White Paper will focus on leaching and permeation of organic contaminants and other metals.

1.1   Permeation of Piping and Non-Metallic Joints

Permeation is a physicochemical mass transfer phenomenon involving diffusion of a solute through a porous medium. The driving force for mass transfer is the presence of an activity (e.g., concentration) gradient with respect to the solute. The rate of permeation can be generalized in simple mathematical terms shown in Equation 1.

(1) $N = UA \cdot (\Delta a)$

where: N = Mass Permeation Rate
                U = Overall Mass Transfer Coefficient
                A = Transfer Area
                a = Solute Activity

The overall mass transfer coefficient (U) is a complex function of the following variables:

- Solute properties (composition, phase)
- Medium properties (composition, pore structure, swollenness)
- Solute-medium interaction (equilibrium partitioning, diffusion coefficient)
- Pipe flow hydrodynamics (Reynolds number)
- Transfer geometry (medium thickness)
- Environmental conditions (temperature)

Permeation of potable water mains and distribution system fittings by external contaminants can be viewed as a three-step process. First, the solute partitions between the external bulk phase (e.g., pore water, soil) and the pipe wall exterior. Next, the solute diffuses through the pore structure of the pipe or fitting. Finally, upon penetration the solute partitions between the internal bulk phase (e.g., pipe water) and the pipe wall interior.

Permeation can occur either from the vapor or aqueous phase. With respect to permeation of potable water mains, the contaminants of interest include highly volatile hydrocarbons and organic solvents. Therefore, both water mains and fittings installed in the vadose and saturated zones are susceptible to contamination by permeation (DWI0441, 1992).

1.2   Metals and Chemical Leaching

Leaching is a broad category that includes the dissolution of a variety of metals and chemicals into drinking water. In some instances, it is difficult to differentiate between corrosion and leaching. Studies have been conducted to determine the rate and extent of leaching from metallic, plastic, and concrete pipes, as well as various coatings, linings, and sealants. Coatings and linings are often employed to prevent corrosion of water mains and mitigate red water problems. Among the more common linings are epoxy resins, cement-mortar, asphalt (bituminous), and concrete.

2.0   Description of Potential Water Quality Problems

Table 1 provides a summary of potential water quality problems associated with permeation and leaching.

Table 1
Summary of Potential Water Quality Problems

Permeation	Leaching
Increased VOC content of distributed water[2]	Increased lead and copper levels[1]
Vinyl Chloride formation[2]	Increased asbestos levels[1]
Aesthetic issues (taste, odor, film formation)	Increased organic contaminants from PE pipe[2]
	Increased metals levels from cement pipe or linings[2]
	Increased organic contaminants from organic linings[2]
	Aesthetic issues (taste, odor, color)

(1) Health effects associated with these parameters have already been addressed by USEPA through existing Safe Drinking Water Act Regulations.
(2) Potential direct public health impact.

The following discussion focuses on the issues listed in Table 1 that can directly impact public health (denoted by a number 2) but have not been addressed through existing Safe Drinking Water Act Regulations within the distribution system.

2.1 Permeation Occurrences and Health Impacts

2.1.1 Dilution Effect

The movement of water through mains acts to dilute contaminants that have permeated the pipe wall. In a simplified hypothetical model involving clean water flowing unidirectionally through a pipe section surrounded by contaminated media (of uniform activity), the solute activity in the pipe water is related to the flow rate (Q) according to equation 2. This is referred to as convective dilution.

(2) $a \propto 1 - e^{(-UA \cdot Q^{-1})}$

The rate and extent of permeation is greatest for small-diameter mains and service lines (DWI0772, 1997). These water lines contain the highest ratio of mass transfer surface area to pipe volume, and are often associated with stagnant or low flow conditions (poor convective dilution). This effect is exacerbated by the greater likelihood of accidental releases of organic contaminants such as petroleum products on a customer's property and consequently, closer to the point of withdrawal or consumption.

2.1.2 Case Studies

More than 100 incidents of drinking water contamination resulting from permeation of subsurface mains and fittings have been reported in the United States (Glaza and Park, 1992). The majority of these incidents were associated with gross soil contamination in the area surrounding the pipe. The occurrence of permeation incidents was equally split between high-risk locations such as: industrial areas; former sites of fuel stations; and near underground storage tanks; and low-risk locations such as residential areas. The sources of contamination for the low-risk areas included disposal and accidental leaking of gasoline, oil, and paint thinner products (Holsen et al., 1991a).

Figure 1 illustrates the distribution system materials involved in the reported permeation incidents. Pipes composed of polymeric materials (i.e., plastics) were involved in 98% of the incidents. These materials include polybutylene, polyethylene, polyvinyl chloride (PVC), and acrylonitrile-butadiene-styrene (ABS). No reported incidents of permeation through metal-based pipe were identified.

Figure 2 illustrates the contaminants involved in reported incidents. The contaminants most likely to permeate plastic are lipophilic and non-polar in nature. Diesel and petroleum products (gasoline-range organics) were involved in 89% of the incidents, while volatile chlorinated solvents accounted for 5% of the incidents. Other contaminants that exhibit high rates of permeation include (simple) chlorinated aromatics, chlorinated and unchlorinated straight-chain aliphatic hydrocarbons, and phenolic compounds (Holsen et al., 1991a; Holsen et al., 1991b). Strongly polar pesticides (e.g. paraquat, malathion, and atrazine) and long-chained (high molecular weight) hydrocarbons were not permeation threats (DWI0032, 1990; Park et al., 1991).
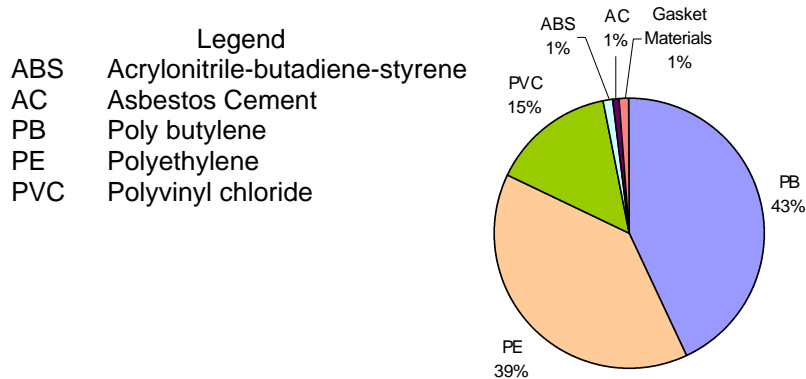
Legend

ABS	Acrylonitrile-butadiene-styrene
AC	Asbestos Cement
PB	Poly butylene
PE	Polyethylene
PVC	Polyvinyl chloride

Figure 1
Pipe materials involved in U.S. water system permeation incidents (Holsen et al., 1991a)

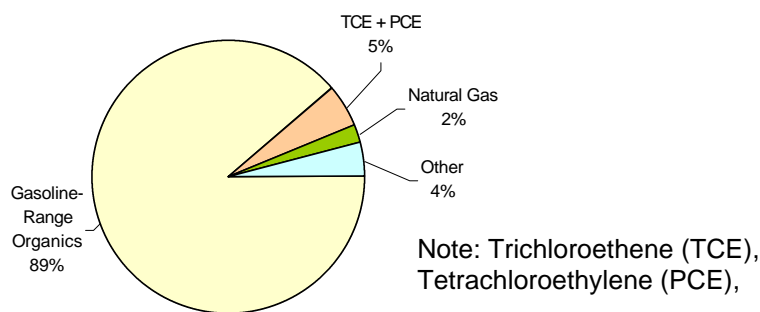Note: Trichloroethene (TCE),
Tetrachloroethylene (PCE),

Figure 2
Contaminants involved in U.S. water system permeation incidents (Holsen et al., 1991)

The occurrence of contamination was generally identified by the customer, and indicated by an unusual taste and odor in the tap water. For many highly toxic substances incorporated in Figure 2, including benzene, vinyl chloride, and dichloromethane, the taste and odor threshold is well above the drinking water Maximum Contaminant Level (MCL) (DWI0441, 1992; Glaza and Park, 1992).

Holsen et al. (1991a) calculated the frequency of permeation incidents for both service connections (Table 2) and water mains (Table 3). The results further indicate that polymeric pipelines are most susceptible to permeation.

Table 2
Frequency of Permeation of Service Connections
(incidents per 10^6 connection-year)

Metal	Polyethylene	Polybutylene	Polyvinyl Chloride
0	3.6	16.5	2.2

Table 3
Frequency of Permeation of Water Mains
(incidents per 10^5 mile-year)

Metal	Concrete	Asbestos Cement	Polyvinyl Chloride
0.1	0	0.3	4.6

Holsen et al. (1991a) conducted an investigation of seven sites where plastic pipe permeation had been reported. All seven incidents involved polybutylene and polyethylene service connections and were associated with gross contamination of the soil surrounding the pipe. In one instance where the cause of contamination was determined to be a 10-gallon gasoline leak onto the road surface, water quality samples were collected from the service line (after 40 hours of stagnation) and analyzed for benzene, toluene, ethylbenzene, and xylene (BTEX). The results, which are presented in Table 4, indicate that the concentrations reached were higher than the numeric value of two drinking water MCLs (MCL violation would be based on annual average exceeding numeric value).

Table 4
Analytical Results from a Gasoline Permeation Incident

Contaminant	MCL (μg/L)	Water Sample (μg/L)
Benzene	5	1,300
Toluene	1,000	4,300
Ethylbenzene	700	< 500
Xylene	10,000	< 500

In March 2000, the Montana DEQ presented water quality results from another incident involving gasoline-contaminated soil. Benzene permeated a 30-inch black polyethylene service line to a concentration of 527 μg/L, over 100 times the drinking water MCL. The contact time between the pipe and contaminant was unknown.

Selleck and Marinas (1991) identified another permeation incident involving a polybutylene service connection. In this case, soil adjacent to the connection had been contaminated with chlorinated aromatics. The results of soil and service line water samples, which are provided in Table 5, indicate that contaminants were capable of permeating to concentrations above health action levels.

Table 5
Analytical Results from a Solvent Permeation Incident

Contaminant	Level of Concern (μg/L)	Water Sample (μg/L)	Wet Soil Sample (g/kg)
1,2-Dichlorobenzene	600[1]	2,500	< 0.010

(1) Numeric value of drinking water MCL; compliance with MCL is based on annual average.

2.1.3 Laboratory Studies

Laboratory studies have been conducted to determine the rate of permeation and time to penetration for several different combinations of contaminants and pipe materials. The primary limitation of these studies is the use of stagnant water columns, and the corresponding lack of convective dilution.

Park et al. (1991) presented the results of pipe-bottle studies using various mixtures of organic chemicals at varying activities. The findings (Table 6) support an important conclusion about the synergistic effects of organic mixtures. The addition of a readily permeable organic chemical to a mixture of relatively non-permeable organic chemicals increases the overall rate of permeation. This consideration is important because most chemical spills and contamination events involve mixtures of similar components (Glaza and Park, 1992).

Table 6
Penetration Times for Organic Solutions Through Polybutylene Pipe

Contaminant	Relative Concentration	Penetration Time[1] (days)	
		Pure	Mixture
Toluene	0.26	38 – 60	16
1,2-Dichlorobenzene	0.19	> 140	16
Toluene	0.24	40 – 60	9
Trichloroethylene	0.21	20	9
Trichloroethylene	0.31	9	9
1,2-Dichlorobenzene	0.24	> 80	10

(1) Defined as the time required to reach 1 mg/L in the pipe water

Joint gaskets have a high intrinsic permeability, but usually are not the primary pathway for permeation. The most common gasket materials include styrene-butadiene-rubber (SBR), chlorinated rubbers, fluorinated rubbers, nitrile rubbers, and ethylene-propylene-diene-monomers. Gaskets and seals are routinely used to join both iron and plastic pipes. According to Park et al. (1991), organic chemicals are approximately 5 to 100 times more permeable in gasket materials compared to polybutylene pipe. However, there are two reasons why most permeation events do not involve gaskets. First, the mass transfer area associated with gaskets is considerably smaller than that associated with pipelines (DWI0772, 1997). Second, gaskets are usually installed in areas where flow is continuous and flow velocities are high, which increases the dilution effectiveness (Holsen et al., 1991a).

2.1.4 Desktop Studies

The large pool of variables and boundary conditions complicate the task of quantifying pipeline permeation. Selleck and Marinas (1991) developed and presented analytical solutions for the permeation of hydrophobic contaminants through plastic water mains. The following two boundary conditions were considered:

Case 1.   New pipe installed in soil subject to gross contamination
Case 2.   Previously contaminated pipe flushed with clean water

The analytical solutions were used to calculate penetration times for ¾-inch polybutylene water mains exposed to a variety of organic contaminants. The results are presented in Table 7. The penetration times for the previously contaminated pipe were small, on the order of minutes. Conversely, the penetration times for the new pipe were on the order of weeks. This phenomenon has been laboratory-verified for PVC and polyethylene pipes as well. Organic chemicals and solvents promote swelling of polymeric materials, which in turn increases the rate of diffusion by several orders of magnitude (Selleck and Marinas, 1991).

Table 7
Penetration Times[1] for Polybutylene Service Lines

Contaminant	Case 1[2]	Case 2[3]
Toluene	36 days	54 minutes
1,2-Dichlorobenzene	53 days	20 minutes
1,3-Dichlorobenzene	65 days	24 minutes
Tetrachloroethylene[5]	13 days	5.5 minutes

(1) Time to: toluene $\geq$ 100 µg/L, 1,2-dichlorobenzene $\geq$ 10 µg/L, 1,3-dichlorobenzene $\geq$ 10 µg/L, tetrachloroethylene $\geq$ 5 µg/L
(2) New pipe
(3) Previously contaminated pipe flushed with clean water

These results indicate that a contamination incident cannot be corrected by simply flushing the line with clean water for a protracted period of time. Although contaminants may be removed from the internal surface, the pipe will retain its status as a swollen, highly permeable medium (Selleck and Marinas, 1991).

New PVC pipes exhibit lower permeation rates than new polyethylene or polybutylene pipes, primarily due to differences in the material matrices (DWI0772, 1997). PVC is an amorphous glassy polymer, while polyethylene and polybutylene are semicrystalline rubber. At low solute activities, PVC is virtually impermeable (penetration time of 10^5 years). However, when exposed to high activity (e.g., saturated) organic conditions, such as those that would occur during gross chemical spillage, PVC pipe is softened to the point of failure. As a result, permeation rates increase dramatically (Selleck and Marinas, 1991; DWI0772, 1997).

Thermodynamic theory indicates that hydrostatic pressure within the pipeline provides negligible resistance to permeation at the pressure range commonly found in the distribution system (Selleck and Marinas, 1991).

2.1.5   Health Impacts

Many of the contaminants involved in permeation events are regulated by federal and state drinking water standards because of known health risks. These include the Phase I, II, IIb, and V Rules of the Safe Drinking Water Act. Maximum Contaminant Levels have been established

and apply at the point of entry to the distribution system. These water quality standards are established based on risks associated with the ingestion (consumption) pathway and typically reflect health consequences resulting from long-term exposures. However, it is important to note that serious acute and chronic health effects can also occur through other pathways, including inhalation and dermal sorption (direct contact). Examples of non-consumptive water use where inhalation and dermal absorption risks may be incurred including showering and hand washing (Argonne National Laboratory Environmental Assessment Division, 2002). Potential health risks associated with these routes of exposure include skin rashes and respiratory distress.

The Chemical Health Effects Tables (U.S. Environmental Protection Agency, 2002a) provides a summary of potential adverse health effects from high/long-term exposure to hazardous chemicals in drinking water.

2.2 Leaching Occurrences and Health Impacts

ANSI/NSF Standard 61: Drinking Water System Components – Health Effects (NSF International, 2001) establishes minimum health effects requirements for the chemical contaminants and impurities that are indirectly imparted (via leaching) to drinking water from products, components, and materials used in drinking water systems. This standard does not establish performance, taste and odor, or microbial growth support requirements for drinking water system products, components, or materials.

The products and materials covered under ANSI/NSF 61 relevant to this White Paper include protective materials (coatings, linings, solvent additives, etc.), joining and sealing materials (solvent cements, welding materials, gaskets, etc.), and pipes and related products (pipes, tanks, fittings, etc.). The NSF Drinking Water Additives Program started with cooperative agreement from the USEPA in 1985. Thus, there are many distribution system materials and components that were installed prior to the adoption of NSF standards. Despite the availability of NSF-approved materials and standards, it is not possible to ensure continuous adherence to existing standards under all circumstances.

A limited survey of 7 states (OR, CA, CO, FL, WA, TX, and NY) suggests that NSF Standard 61 certification is commonly required by State Health Departments for the components of drinking water distribution systems. All seven of the primacy agencies interviewed mandate the use of materials approved under NSF 61.

The overall and general method for certifying a particular product is described herein. First, product-specific information is reviewed "to determine the appropriate analytical testing and to ensure that the potential health effects of products or materials are accurately and adequately identified". This information includes but is not limited to complete formulation information for each water-contact material (e.g. percent or parts by weight for each chemical in the formulation), the total volume of water that the product can hold when filled to capacity, the maximum temperature to which the product, component, or material is exposed during its intended end use, and a list of published and unpublished toxicological studies relevant to the chemicals and impurities present in the product, component, or material.

Appropriate analytical testing of the exposure water includes a "minimum test battery" established for a material of common formulation and/or formulation-dependent analysis for uncommon or non-standard formulations.

The extraction procedures used to generate exposure water varies by product category (e.g. pipes and related products, barrier materials, joining and sealing materials, mechanical devices and others), by application (hot or cold etc.) and by configuration, size and material composition of the individual product as applicable. Some of the variables to be selected and/or adjusted include surface area-to-volume ratio, conditioning (e.g. cleaning and pre-soaking), exposure time, and single vs. multiple time point sampling protocols, filling, emersion or other contact arrangement and initial test-water quality characteristics.

Four extraction waters are specified and available for exposure:

(1) pH = 5, with 2 mg/L available chlorine and 100 mg/L hardness;
(2) pH = 6.5, with 2 mg/L available chlorine and 100 mg/L hardness;
(3) pH = 8 (organic analysis), with 0 mg/L available chlorine and 100 mg/L hardness;
(4) pH = 10, 2 mg/L available chlorine.

Methods of preparing these waters are specified and the water or waters to be used for testing are either specified by material (e.g. the pH 6.5 water and the pH 10 water for copper and copper alloy pipe and tubing), or by formulation-specific selection criteria. Several entries within the standard direct the use of a conservative approach, for example:

> "Products or materials whose intended uses fall under more than one section of this Standard shall be evaluated under the section having the most rigorous evaluation conditions. NOTE – Rigorous conditions are typically associated with shorter conditioning periods, longer exposure periods, higher surface-to-volume ratios and higher exposure temperatures."

The results of the laboratory analysis on the exposed extraction water are normalized to determine the level of contaminants projected "at the tap". The normalized contaminant concentrations are then compared to various criteria. The water quality criteria used to evaluate the normalized concentrations come from several sources. There are normative criteria based on the contaminants regulated by the USEPA and established by Health Canada or addressed by USEPA guidance. The single product allowable concentration (SPAC) utilized by NSF Standard 61 is one tenth of the regulatory (or guidance) MCL (or MAC), except for radionuclides for which the values are the same. There are six additional substances for which a SPAC has been established by NSF International and peer-reviewed. There are over 300 additional substances listed in NSF Standard 61 as threshold-of-evaluation chemicals, for which too little data exists to determine a SPAC and for which "a comprehensive literature search for the particular substance and consideration of structure-activity relationships" must be undertaken if certain threshold concentrations are exceeded.

NSF 61 also applies "informational drinking water criteria" which includes over 60 additional compounds with SPACs that have not been peer reviewed.

2.2.1   Plastic Water Mains

Polyvinyl chloride (PVC) mains manufactured prior to 1977 contain elevated levels of vinyl chloride monomer, which are prone to leaching (Flournoy, 1999). Vinyl chloride is a toxic chemical with known carcinogenic effects; the drinking water MCL is 2 µg/L, enforceable at the point of entry to the distribution system.

Water quality samples collected from a rural water system in Kansas, which had installed over 100 miles of pre-1977 PVC, contained as much as 14 µg/L of VC. Of the 53 samples collected (from 4 unique sites) over a 6-year period, 55% exceeded the VC MCL. This finding motivated other states with pre-1977 PVC to monitor for VC within the distribution system. Other VC MCL exceedances were identified in Missouri, Iowa, Arkansas, and Texas. The study identified the following factors which promoted VC leaching:

- Small-diameter pipes (i.e., ≤ 2 inches)
- High temperature (i.e., ≥ 50 ^{o}F)
- High contact times with pre-1977 PVC pipe (i.e., ≥ 1 day)

No instances of MCL violations associated with leaching from PVC mains manufactured post-1977 were cited in the literature. Additional research is required to estimate the quantity of pre-1977 PVC mains still in use.

According to "The Handbook of PVC Pipe" (Uni-Bell, 3rd Edition) PVC pipe and fittings are immune to nearly all types of corrosion that could result in the leaching of corrosion products. The Handbook provides detailed data regarding the resistance of PVC pipe to numerous chemicals and states that PVC pipe and fittings are resistant to chemicals generally found in water and sewer systems.

Advantages and drawbacks of using plastic water mains are summarized in Table 8.

Table 8
Features of Plastic Pipe

Advantages	Drawbacks
• Resistant to chemical corrosion and tuberculation	• Vulnerable to permeation by organics
• Do not require linings	• May leach organic chemicals
• Low relative cost	• More frequent mechanical failure
• Ease of handling and installation	• Reduced strength and flexibility (kinking)

2.2.2   Cement Materials

Cement-based materials include reinforced or pre-stressed concrete pipes, cement-mortar linings, and asbestos-cement pipe. Two general components of cement-based materials include the aggregates and the binder. The binder consists of calcium silicates and calcium aluminates in various proportions depending on the type of the cement (Leroy, Schock, Wagner, and Holtschulte, 1996).

Several types of degradation of cement materials can occur in the presence of acid waters or waters aggressive to calcium carbonate (Leroy, Schock, Wagner, and Holtschulte, 1996). Waters that have a very low ion content are aggressive to calcium hydroxide contained in hydrated cements (ACIPCO, 2002). These waters will also attack calcium silicate hydrates, which form the larger portion of cement hydrates. Although calcium silicate hydrates are nearly insoluble, soft waters can progressively hydrolyze them into silica gels, resulting in a soft surface with reduced mechanical strength. Calcium hydroxide will also leach from cement-mortar linings exposed to soft waters. The extent of leaching increases with the aggressiveness of the water and its residual time in the pipe and is inversely proportional to the diameter of the pipe.

Douglas and Merrill (1991) studied the deterioration of new cement-mortar linings under various water quality conditions. Field-testing demonstrated that aggressive water is capable of leaching cement compounds from these linings, causing significant increases in bulk solution pH, alkalinity, and calcium. The pH of stagnant water held in contact with cement-mortar lined pipes for one week increased from an initial value of 7 upwards as high as 12. While convective dilution reduced the magnitude of these water quality changes, it also resulted in a greater overall extent of leaching, as indicated by the depth of the linings calcium loss. Asphaltic seal coats significantly reduced cement-mortar deterioration.

Cement materials contain a variety of regulated inorganic chemicals, many of which are prone to leaching. Guo et al. (1998) conducted laboratory tests to determine the extent of leaching from ductile iron pipes lined *in situ* with portland cement (type I) mortar. The pipes were lined and cured in accordance with ANSI/AWWA Standard C602-89, and subsequently disinfected according to ANSI/AWWA C651-92. The test water was standard faucet water from a New Jersey utility. Under static conditions, arsenic, barium, cadmium, and chromium leached from the lining to maximum values roughly 10-20% of their respective drinking water MCLs. Further, the acid-soluble contents of arsenic, barium, cadmium, and chromium in the cement coating applied were only 3, 1.9, 13, and 6.6 percent of the cement industry maximum, respectively. Therefore, the extent of leaching could have been higher if an alternate cement lining had been applied. Additional research is necessary to understand the fate of heavy metals associated with cement leaching, and the degree of accumulation within distribution systems.

According to Berend and Trouwborst (1999), the application of cement-mortar lining can also lead to aluminum leaching. The aluminum content in cement-mortar linings varies, as shown in Table 9. One of the predominant crystalline phases in cement is tricalcium aluminate, which is believed to dissolve according to the following reaction:
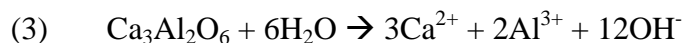
(3) $Ca_3Al_2O_6 + 6H_2O \rightarrow 3Ca^{2+} + 2Al^{3+} + 12OH^-$

Table 9 Aluminum Composition of Common Cement Linings (% as Al_2O_3)				
Blast Furnace Cement	**Pozzolanic Metallurgical Cement**	**High-Alumina Cement**	**Portland Cement**	**Berend and Trouwborst Study Cement**
13%	13%	36%	5%	18.7%

Aluminum poses serious health risks to hemodialysis patients. The European Union defines a maximum aluminum concentration of 30 µg/L in water used for hemodialysis. The USEPA has established a Secondary MCL range for aluminum of 50-200 µg/L.

In the study by Berend and Trouwborst (1999), the installation of 7,200 feet of cement-mortar lined ductile iron pipe caused aluminum levels in a water supply to increase from 5 µg/L to 690 µg/L over the course of 2 months. More than two years later, aluminum continued to leach from the lining and produce water with over 100 ìg/L of aluminum. This was attributed to several illnesses and a 32% mortality rate at a receiving dialysis center. The water in contact with the pipe was seawater that had been desalinated and subsequently treated with coal-filtration, fluoridation, and UV disinfection. The water was aggressive (maximum Langelier Index between –0.5 and –1.5), soft (hardness 15-20 mg/L as $CaCO_3$), of low alkalinity (no data) and high pH (8.5 to 9.5). The pipe had been lined with cement mortar at the factory by a rotary centrifugal process.

Aggressive, soft, and poorly buffered (i.e., low alkalinity) waters promote aluminum leaching from cementitious materials. These are the same water quality conditions that are conducive to leaching of lead and copper. The Lead and Copper Rule (USEPA, 1991) specifies a minimum pH within the distribution system of 7.0, but does not specify minimum alkalinity levels. Utilities are required to maintain optimal water quality parameters at the point of entry to the distribution system and at several locations within the distribution system to minimize lead and copper leaching at the tap.

The extent of leaching is also strongly related to the contact time between the water and the cement-mortar lining. In the study by Berend and Trouwborst (1999), the average contact time was 2.3 days.

Polyphosphate corrosion inhibitors attack and soften cement linings, thereby accelerating cementitious leaching. These corrosion inhibitors can also chelate and complex with soluble calcium and aluminum.

2.2.3 Bituminous Coatings and Linings

Asphalts and coal tars are bituminous materials that occur naturally or are derived from non-destructive separation of petroleum fractions. Bituminous coatings contain a complex mixture of many chemical compounds, the exact composition of which depends upon the individual source. Historically, the use of bituminous coatings has resulted in problems when exposed to soft,

acidic waters. Among these problems are red water discoloration, extensive tuberculation, and loss of carrying capacity.

The material properties of the coating systems are enhanced by the use of additives, the most obvious of which are the solvents used as a vehicle for application. However, these additives can diffuse through the coating and into the pipe water. Elevated levels of alkyl benzenes and polycyclic aromatic hydrocarbons (PAHs) have been reported in reservoirs with new bituminous coatings and linings (Yoo et al, 1984; Krasner and Means, 1985; Alben, 1980). While few additives from coating materials are included in prioritized lists of SOCs to be monitored in drinking water, proper application, curing, and testing prior to returning coated surfaces to service will minimize subsequent exposure to solvents used in application.

Alben et al (1989) studied leaching of organic contaminants from flat steel panels lined with various coatings, including vinyl, chlorinated rubber, epoxy, asphalt, and coal tar. Emphasis was given to the rate of leachate production and leachate composition. The test water was GAC processed tap water with a pH of 8 to 9. Leaching rates (mg/m^2-day or µg/L-day) were assessed over a period of 30 days. Organic contaminants were found at parts-per-billion levels in water compared to parts-per-thousand levels in the coating.

Concentrations of total organic carbon (TOC) in leachate from asphalt were the lowest of the five coatings studied. Alkyl benzenes constituted 72% of the leachate from asphalt coating. Based on the results of 30-day laboratory experiments with asphalt-coated test panels (coating thickness 536 µm), the rates of leaching were predicted for full-scale storage tanks with volume to surface area ratios ranging from 8.2 to 37.7 ft^3/ft^2 (2.5 to 11.5 m^3/m^2). Initial rates of leaching for the semi-volatile base-neutrals were found to range from 0.4 to 1.7 µg/L-day; after 30 days the rates dropped to 0.2 to 1.0 µg/L-day.

Concentrations of TOC in leachate from coal tar were the second lowest of the five coatings studied (2 bituminous and 3 polymeric coatings). PAHs accounted for essentially all of the TOC in leachate from coal tar coating. Based on the results of 30-day laboratory experiments with coal tar-coated test panels (coating thickness 2,746 µm), the rates of leaching were predicted for full-scale storage tanks with volume to surface area ratios ranging from 8.2 to 37.7 ft^3/ft^2 (2.5 to 11.5 m^3/m^2). Initial rates of leaching for the PAHs were found to range from 2.6 to 12 µg/L-day; after 30 days the rates were similar at 2.3 to 11 µg/L-day.

There have been incidents of PAH leaching from coal tar linings that have ultimately led to taste and odor complaints. In one particular case, a butterfly valve failed while in the closed position, thus creating an artificial dead-end along a pipe stretch coated with coal tar. Upon repairing the valve and reopening it, the utility received several odor complaints. Water quality sampling revealed that anthracene, naphthalene, and other PAHs were present at concentrations in excess of 5,000 µg/L (Satchwill, 2002).

2.2.4   Epoxy Coatings and Linings

Epoxy coating starts out as a dry powder which is produced by combining organic epoxy resins with appropriate curing agents, fillers, and pigments. When heated, the powder melts and its constituents react to form complex cross-linked polymers.

Alben et al (1989) studied leaching of organic contaminants from epoxy-coated flat steel panels, with emphasis on the rate of leachate production and leachate composition. The test water was GAC processed tap water with a pH of 8 to 9. Leaching rates (mg/m^2-day or $\mu g/L$-day) were assessed over a period of 30 days and found to decay as a simple exponential.

Concentrations of TOC in leachate from epoxy resin were the highest of the five coatings studied (2 bituminous and 3 polymeric coatings). The solvents methyl isobutyl ketone (MIBK), ortho-, meta-, and para-xylene represented a major portion (51%) of leachate from the epoxy coating. Based on the results of 30-day laboratory experiments with epoxy-coated test panels (coating thickness 393 μm), the rates of leaching were predicted for full-scale storage tanks with volume to surface area ratios ranging from 8.2 to 37.7 ft^3/ft^2 (2.5 to 11.5 m^3/m^2). Initial rates of leaching for the solvents were found to range from 40 to 187 $\mu g/L$-day; after 30 days the rates dropped to 8 to 37 $\mu g/L$-day.

These findings were further supported by field studies. Solvents (MIBK, xylenes) from an epoxy coating were also detected in effluent from 2 of 3 storage tanks that were monitored one month after application of the coating (Alben et al, 1989).

The extent of leaching of organic contaminants from epoxy resin linings was found to be strongly dependent on the duration of the curing process (DWI0032). Longer curing periods produced more stable linings.

The City of Calgary performed leaching studies with pipe coupons coated with two-component epoxy resins (Satchwill, 2002). Five different NSF and AWWA approved epoxy resins were independently investigated. Application and curing of the epoxy resin involved the following steps: (1) applying the initial coating followed by verification that application meets wet film thickness specification, (2) 24 hours of air drying followed by verification that application meets dry film thickness specification, (3) applying a second epoxy coating (followed by verification), and (4) 24 hours of air drying (followed by verification.) The coupons were then filled with water and held for 72 hours prior to analysis for TOC and VOCs. The results, which are provided in Table 10, illustrate the extreme extent of leaching from new epoxy resins into stagnant water columns.

Table 10
72-Hour Epoxy Resin Leaching Results

Coating	BTEX (mg/L)	TOC (mg/L)
Epoxy 1	13.2	166
Epoxy 2	4.9	34
Epoxy 3	48.0	279
Epoxy 4	0.2	345
Epoxy 5	25.6	143

Benzyl alcohol and epoxy resin oligomers have been identified in leachate from epoxy coated water mains (Crathorne et al, 1984; Watts et al, 1983).

3.0   Prevention and Mitigation Methods

3.1   Indicators

The majority of the contaminants of concern with respect to permeation and leaching events are organic chemicals, including volatile, semi-volatile, and non-volatile substances. While many of these contaminants are regulated under national and/or state drinking water standards, and are thus monitored at the source, others fall into the class of unregulated organics. Some of these unregulated contaminants are potential health concerns and are listed on the Contaminant Candidate List (CCL) under the Unregulated Contaminant Monitoring Rule (UCMR).

Source water quality monitoring will not aid in detecting contamination events that occur within the distribution system, including permeation and leaching. However, there are other methods and indicators that can be used to prevent and identify contamination events. The following potential indicators are addressed:

- Taste and odor of tap water
- Installation inspection/Site use review
- Environmental monitoring
- Release to service monitoring

3.1.1   Taste and Odor of Water

Taste and odor (T&O) complaints have previously led to the identification of gross subsurface contamination. The key question in this regard is whether or not T&O detection is useful for identifying a contamination event *before* significant health risks are incurred (through consumption). Table 11 provides a comparison of risk thresholds versus T&O thresholds for several organic substances commonly involved in permeation and leaching events. The effect of mixing chemicals on the T&O thresholds is unknown.

Table 11
Organoleptic versus Risk Thresholds for Pure Compounds

Contaminant	MCL	Taste Threshold	Odor Threshold	Detectable Before Risk? (3)
Benzene	5 µg/L	500-4,500 µg/L	2,000 µg/L	No
Ethylbenzene	700 µg/L	29 µg/L	29 µg/L	Yes
Xylenes	10 mg/L	No Data	2.2 mg/L	Yes
Naphthalene	300 µg/L[1]	No Data	300 µg/L	No
1,2-Dichloroethane	5 µg/L	29,000 µg/L	No Data	No
Tetrachloroethylene (PCE)	5 µg/L	300 µg/L	No Data	No
Methyl Isobutyl Ketone (MIBK)	500 µg/L[2]	No Data	20,000 µg/L	No
Vinyl Chloride	2 µg/L	None	None	No
Styrene	100 µg/L	730 µg/L	No Data	No

(1)   State of New Jersey MCL
(2)   State of Wisconsin MCL
(3)   Health risk level based on long-term exposure.

In most instances, the risk threshold is substantially lower than either the taste or odor thresholds. However, while there are contaminants for which the organoleptic thresholds are lower than the levels where long-term exposure is a concern, the most prevalent sources of contamination are events like petroleum product releases where a combination of compounds with varying organoleptic and risk thresholds will be present.

3.1.2   Installation Inspection/ Site Use Review

There are currently no certification requirements in published literature for installers or inspectors with regard to identifying potential subsurface contamination or installing pipes or fittings in regions of potential contamination. Standards ANSI/AWWA C900 through C950 cover plastic pipe and recommend consultation with the pipe manufacturer prior to selecting a material if a water main must pass through an area of gross contamination. The UPC specifies that clean soil be used as backfill around buried pipes.

Activities and procedures that are typical of construction within areas believed to be at significant risk of contamination include reviewing site use(s) and performing environmental monitoring during installation.

Historical and current site use will allow the construction team to assess the probability of contamination as well as the specific contaminants for which to look. The contamination of soil and/or groundwater by highly permeable substances can often be identified during construction activities through the use of air quality monitoring equipment and/or passive soil gas surveys. Gas chromatographic instruments, including photo-ionization detectors (PIDs) and flame-ionization detectors (FIDs), are tools that can be used to detect hydrocarbons in the vapor phase (or headspace) at the parts-per-million level. Passive soil gas sampling involves the use of specially designed sorbents to capture volatile and semi-volatile organic compounds in soil and

water. Passive soil gas sampling provides quantitative, time-integrated data of organic contamination.

3.1.3 Monitoring

Monitoring is critical to ensure that a new or rehabilitated main can be safely released to service. Some utilities have developed water quality "release to service" standards for new and repaired mains. Typical monitoring parameters include total coliform bacteria, HPC, pH, disinfectant residual, turbidity, and odor. However, monitoring for additional parameters may be warranted based on knowledge of materials used and/or site-specific water quality or environmental conditions. For example, monitoring for BTEX and MIBK following application of an epoxy coating can be used to evaluate the extent of leaching and potentially identify problems with the lining material or manufacturers defects.

3.2 Maintenance

Stagnation of water can exacerbate permeation and leaching incidents. Poor convective dilution of permeated or leached solvents is associated with stagnant areas of the distribution system. The extent of leaching of cementitious and organic material exhibits some relationship to the contact time between the pipe and internal water. Unidirectional flushing and reservoir turnover can be used to encourage fluid movement, minimize residence time, and replace stagnant water. Flushing does not suppress the process of leaching, but the movement of fluid helps to prevent the accumulation of contaminants in a localized area. Studies have demonstrated the ineffectiveness of flushing to mitigate a permeation incident. While flushing may replace the contaminated water, the pipeline retains its swollen, highly permeable state. In these instances, pipe replacement is an effective practice.

With respect to permeation, most incidents occur along service connections due to their small diameter and frequent stagnation, however, service lines are not owned by the utility (USEPA, 1999). Thus, in the event of a permeation event involving a service connection the consumer would need to flush the tap long enough to draw water from a point beyond the service line to avoid exposure to potentially contaminated water.

3.2 Design and Installation

Prevention of permeation and leaching requires proper materials selection and installation practices. The phenomena of permeation and leaching require consideration of the environments surrounding and within the pipe, respectively. From a design perspective, permeation can be precluded by identifying the area of installation and recognizing the potential for past or future gross spillage of organic chemicals and petroleum products.

ANSI/AWWA Standards C900 through C950 state that if a water main must pass through an area of gross contamination, the manufacturer should be consulted regarding the permeation of pipe walls and joint fittings prior to selecting the material. The Standards also note that research has documented that pipe materials such as polyethylene, polybutylene, polyvinyl chloride, and asbestos cement; and elastomers, may be subject to permeation by lower molecular weight

organic solvents or petroleum products. Ten State Standards (Great Lakes…1997) states that non-permeable materials shall be used for all portions of the system including water mains, service connections, and hydrant leads in areas of distribution systems where the groundwater is contaminated by organic compounds. The Uniform Plumbing Code does not discuss permeation directly. It does state that clean soil will be used as backfill around buried pipes and seems to recognize some potential for contamination from the pipe-embedment environment   The UPC does not, however, specifically address installation through contaminated soils or permeation as a pathway of exposure.

To prevent leaching from cement-based pipes and linings, knowledge of water quality is necessary when determining the appropriateness of certain materials. ANSI/AWWA Standard C401-98 provides guidance on the application of asbestos cement pipes based on internal water quality characteristics, as summarized in Table 12.

Table 12
Aggressiveness of Water towards Asbestos-Cement Pipe

Water Type	$pH + log(AH)$	Langelier Index	Recommended Pipe Type
Highly Aggressive	< 10.0	< -2.0	*
Moderately Aggressive	10.0 – 11.9	-2.0 to –0.1	II
Nonaggressive	≥ 12.0	≥ 0	I and II

Source: ANSI/AWWA Standard C401-98
*The serviceability of pipe for such applications should be established by the purchaser in conjunction with the manufacturer
Type I – no limit on uncombined calcium hydroxide
Type II – 1.0% or less uncombined calcium hydroxide
A = Alkalinity as mg/L $CaCO_3$; H = Calcium hardness as mg/L $CaCO_3$

The rate of leaching of organic additives was found to decrease exponentially with time. Therefore, it is recommended that newly lined pipes be pre-soaked prior to release to service. Normal hydrostatic testing and disinfection activities will help remediate leaching. Extending the curing process will also help improve the stability of epoxy linings.

Additional Standards applicable to design, installation, and application of pipelines, linings, and coatings are summarized below:

- AWWA Manual M9 provides information related to design, handling, delivery, laying, field testing, and disinfection of concrete pressure pipe.
- AWWA Standard C600-606 covers pipe installation procedures
- ANSI/AWWA C104 through C153 covers ductile iron pipe, fittings, linings, coatings, gaskets, and joints.
- ANSI/AWWA C200 through 222 covers steel pipe, linings, coatings, and joints.
- ANSI/AWWA C300 through 304 covers concrete pipe.
- ANSI/AWWA C400 through C403 covers AC Pipe.
- ANSI/AWWA C800-89 covers service lines.

3.3 Source Water Treatment

Water quality plays in important role in the process of materials leaching. Key parameters in this regard include pH, alkalinity, and dissolved inorganic ions. Water quality parameter requirements for minimizing soluble lead and copper levels are provided in the Lead and Copper Rule (USEPA, 1991). Suitable water quality conditions for the installation of asbestos-cement pipe were summarized previously.

Based on a study by Brend and Trouwborst (1999), aluminum leaching from portland cement and blast furnace slag cement only occurred in low alkalinity water. Based on this study, the following recommendations were established:

- In-situ linings of ordinary portland cement should not be used if the alkalinity is less than 55 mg/L as $CaCO_3$.
- Factory-applied cement linings should not be used if the alkalinity is below 25 mg/L as $CaCO_3$.

Vik and Hedberg (1991) provided the following recommended water quality conditions to avoid leaching of cement-based materials, including AC and concrete pipes and cement mortar lining.

- pH > 7
- Alkalinity > 15 mg/L as $CaCO_3$
- Calcium > 10 mg/L
- Sulfate < 200 mg/L
- Aggressive CO_2 < 5 mg/L

4.0 Summary

Distribution system infrastructure and appurtenances including piping, linings, fixtures, and solders can react with the water they supply as well as the external environment. Permeation and leaching are two mechanisms that can result in the degradation of the distributed water. Leaching from cement linings can occur in soft, aggressive, poorly buffered waters. Under static conditions, metals such as aluminum, arsenic, barium, chromium, and cadmium can leach from cement linings, even when NSF approved materials are used and linings are applied according to AWWA standards. Current provisions of the Safe Drinking Water Act do not require monitoring for heavy metals beyond the point of entry to the distribution system, and additional research would be required to assess the degree of metals accumulation within the distribution system. Vinyl chloride can leach from pre-1977 PVC pipe. No instances of MCL violations were cited in association with post 1977 PVC pipe. Volatile organics present in the ground water or vadose zone can permeate plastic piping and gaskets. Permeation is typically most severe for small diameter, low-flow pipes. The literature cites instances where MCL violations have occurred at the point of consumption, although current provisions of the Safe Drinking Water Act do not require monitoring for volatile organic compounds beyond the point of entry to the distribution system. In most instances, the risk threshold of chemical contaminants is substantially lower than either the taste or odor thresholds, suggesting that utilities cannot rely

confidently on customers' perception of taste and odor for identifying contamination events. Unidirectional flushing can be used to rid the distribution system of stagnant, contaminated water, but additional research is needed to determine the fraction of heavy metals and organics that can be removed through flushing. Permeated plastic piping must be replaced since the piping retains its swollen porous state after permeation. NSF Standard 61 and numerous AWWA Standards have been developed to prevent the degradation of drinking water due to contact with piping materials. Materials selection, design, and installation considerations based on water quality and environmental conditions are addressed in these Standards.

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