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Methodology for scenario-based assessments and demonstration of treatment effectiveness using the Leaching Environmental Assessment Framework (LEAF)

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Abstract

A methodology for developing scenario-based leaching assessments as part of the Leaching Environmental Assessment Framework (LEAF) is illustrated using a hypothetical management/treatment scenario of contaminated soil from a copper and lead smelter. Scenario assessments refine the process beyond screening-level assessments by considering site- and scenario-specific information about the disposal or utilization environment. LEAF assessments assume (i) granular materials leach at local equilibrium with percolating water, while (ii) monolithic materials (e.g., low permeability solidified/stabilized soils) leach by diffusion-based mass transport toward surrounding contact water. Leaching concentrations, estimated using LEAF leaching test data and estimated or measured scenario information, are compared to threshold values. Demonstration results indicate that leaching from untreated soil is significantly ($> 10\times$) greater from solidified/stabilized soil than treated material, except for highly soluble constituents (Cl^- , NO_3^{2-}) or when constituents have similar equilibrium concentrations in both materials (As, Pb). Comparison between wet and dry environments show that while dry environments lead to lower COPC mass-based rates of leaching, the leaching concentrations may be higher due to lower liquid-to-solid ratios. The presented assessment methodology can be used to evaluate treatment effectiveness when both physical and chemical retention characteristics of the material are altered.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRedit authorship contribution statement

Andrew Garrabrants: Conceptualization, Methodology, Writing - original draft. **David Kosson:** Conceptualization, Methodology, Writing - original draft. **Kevin Brown:** Validation, Writing - review & editing. **Daniel Fagnant:** Writing - review & editing. **Gregory Helms:** Writing - review & editing. **Susan Thorneloe:** Writing - review & editing.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2020.124635.

Keywords

Leaching assessment; Percolation; Mass transport; Smelter soil; Solidification/stabilization

1. Introduction

Leaching assessments estimate the release of constituents of potential concern (COPCs) from solid wastes and secondary materials into water resources and ecosystems that may result from contact with water. Typically, leaching assessments are based on single extraction test results, e.g., U.S. EPA's Toxicity Characteristic Leaching Procedure (TCLP, Method 1311) or Synthetic Precipitation Leaching Procedure (SPLP, Method 1312) that assume a leaching condition with little relevance to the anticipated end use (Clavier et al., 2019; da Silva et al., 2017; Intrakamhaeng, et al., 2019; U.S. EPA, 1991, 1999). The Leaching Environmental Assessment Framework (LEAF) was developed to provide a flexible basis for leaching assessments based on measured intrinsic leaching characteristics that can be used to develop more accurate leaching assessments for waste treatment, disposal, use, and remediation scenarios (Kosson et al., 2002; Garraabrants et al., 2010; Kosson et al., 2014; U.S. EPA, 2019a). LEAF test methods (U.S. EPA, 2019b) are designed to characterize for COPCs (i) the liquid-solid equilibrium as a function of pH and liquid-solid ratio (L/S in L/kg-dry), (ii) the elution during percolation under local equilibrium, and (iii) the mass-transfer (diffusion) rate controlling release from low permeability materials (see method summaries in SI-1).

Within LEAF, assessments are derived from material characteristics, leach testing results, and assumptions regarding disposal or use conditions (U.S. EPA, 2014a, 2014b) with the resultant leaching estimates directly compared to acceptance thresholds or used as input to fate and transport or exposure models supporting human health and ecosystem risk assessments. The required refinement of assessment, however, depends on the intended use of the leaching assessment and decision thresholds,¹ while the accuracy of the estimate is constrained by the available input data, methodology, and underlying assumptions. For example, screening-level assessments (Garraabrants et al., 2020) are intended to be bounding, biasing toward over-estimating COPC leaching to allow management decisions to be protective of the environment while balancing simplifications and uncertainty in the assessment approach. Screening-level assessments may be helpful when preliminary estimates of leaching concentrations are desired or when the goal of the assessment is to rank or select relevant COPCs.

However, screening-level estimates lack the level of detail required (i) to evaluate potential leaching for site-specific scenarios, (ii) to provide comparisons between different leaching conditions (e.g., water percolating through granular material versus water flow around monolithic materials), or (iii) to account for material characteristic changes over time

¹Often, numerical threshold criteria (e.g., toxicity characteristic standards or drinking water standards) are established to guide decisions based on leaching assessment results. Specific threshold criteria may be health-based values dependent on applicable regulations of the leaching assessment. If a leaching assessment indicates that projected concentrations are below the threshold using conservative assumptions (biased towards greater estimated leaching for uncertain parameters and scenario assumptions), there most likely will not be a need to refine the leaching assessment using additional site-specific and scenario-specific information.

(Branch et al., 2017; Garraabrants et al., 2015; Kosson et al., 2002, 2014). Scenario assessments refine the leaching assessment process using site- and scenario-specific information on application scale and environmental parameters to provide a more precise estimate, as well as a common basis, for comparison of leaching scenarios with dissimilar conceptual release models (e.g., percolation vs diffusion). The assessment scenario can be designed to reflect a range of anticipated environmental conditions with one or more sets of bounding conditions that account for the hydraulic, physical, and chemical nature of materials.

The Interstate Technology and Research Council (ITRC) recommended a scenario-based assessment approach using LEAF leaching tests for demonstrating treatment effectiveness of solidified/stabilized (S/S) materials (ITRC, 2011). However, the ITRC guidelines focus on release from S/S materials placed within groundwater, whereas literature examples are not widely available for S/S materials placed in the vadose zone.

Scenario-based assessments, extending recent LEAF technical guidance (U.S. EPA, 2019), are presented here using a hypothetical case study of S/S-treated soil from a copper and lead smelter site. These assessments compare estimated leaching from untreated soil and S/S treated material managed above the water table using the same materials, methods, and leaching results used to develop screening-level assessments (Garraabrants et al., 2020). Scenario-specific information, including the physical dimensions of the material, the mode of water contact based on the physical form of the material, and the water infiltration for selected wet and dry environments, was derived from regional information and is not intended to represent any particular application or location. This work illustrates (i) a methodology for developing scenario-based leaching assessments using LEAF testing results in conjunction with scenario-specific information; (ii) the effects of scenario parameters on scenario-based assessment results; and (iii) a pathway for evaluating the effectiveness of treatment.

2. Hypothetical case study

The soil surrounding a copper and lead smelter is contaminated with heavy metals and other COPCs, affecting a 400-m² plan-view area (20 m by 20 m) from grade to a depth of 5 m (total volume of 2000 m³). The soil is a sandy loam with an assumed porosity of 0.4, dry density of 1600 kg/m³, and field capacity of 0.24 (Meyer and Gee, 1999). The water table does not directly contact, or otherwise affect, the source material. The field scenario was assumed oxic (chemically oxidizing or mildly reducing) to match laboratory testing carried out under oxic conditions. The leaching assessment period is 30 years evaluated in 1-year time steps.² For this scenario, threshold concentrations are U.S. national primary and secondary drinking water standards (U.S. EPA, 2009).³

²An assessment duration of 30 years was selected for this illustration because it is the default post-closure care period under RCRA (U.S. EPA, 2016); however, a case-by-case evaluation period may be longer than 30 years.

³In this illustrative scenario, primary and secondary drinking water standards were selected for demonstration purposes only. The selection of acceptable threshold values should be made on a site-by-site basis in consultation with the applicable regulatory authority.

2.1. Study materials

Detailed descriptions of the untreated contaminated field soil (CFS) and S/S-treated contaminated field soil (S/S-CFS) are presented in Garraabrants et al. (2020). Total content analysis of the CFS indicates cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), and zinc (Zn) in excess of 1000 mg/kg. Other COPCs with applicable threshold criteria include arsenic (As), barium (Ba), beryllium (Be), chloride (Cl^-), fluoride (F^-), nitrate (NO_3^-), antimony (Sb), selenium (Se), sulfate (SO_4^{2-}) and thallium (Tl). S/S treatment consisted of mixing CFS with Portland cement (12% by dry mass) and adding water to hydrate the mixture. No attempt was made to optimize the treatment process because the goal was to create a monolithic material to illustrate the assessment process and not to demonstrate the efficacy of any specific treatment.

2.2. Water contact mode and required leaching data

LEAF guidance (U.S. EPA, 2019a) includes a flowchart (Fig. 1) for development of scenario-based assessments dominated by flow-through or flow-around modes of water contact. The water contact mode determines which the leaching data and LEAF methods are required to support the assessment.

As a near surface soil, CFS is compacted such that infiltrating water flows through the soil column creating a percolation scenario. Leachate is in local equilibrium with the soil and at a concentration that can change as minerals dissolve with the cumulative volume of infiltration passing through the soil (Bandow et al., 2019). Therefore, knowledge of COPC equilibrium concentrations over an applicable pH domain (from pH-dependent batch leaching; Method 1313) and evolution of leaching concentrations with continued infiltration (from percolation column leaching; Method 1314) are important for percolation scenarios.

After S/S treatment, the physical form of the soil is altered by the hydration of cement to a lower permeability relative to surrounding soils such that infiltrating water is diverted around the S/S-CFS material. COPCs must diffuse through interstitial pores of the low permeability material to the surrounding infiltrating water. The rate of leaching is controlled by the rate of diffusive mass transport within the monolith as driven by internal concentration gradients between the core and surface of the material. In addition, changes in the composition and geochemistry of the S/S material increase the natural pH of S/S-CFS which dictates the COPC concentration in the internal pore water. For a scenario dominated by the diffusion mass transport rate (subsequently referred to as a “diffusion scenario”), important test data include the pH-dependent equilibrium of COPCs (Method 1313) and the rate of COPC mass transport within the S/S material to the exposed surface (Method 1315).

Example leaching tests results are presented for arsenic and selenium from CFS and S/S-CFS (Fig. 2). Method 1313 data shows batch extraction concentrations in equilibrium with solid material at different target pH values. The natural pH in pH-dependent leaching tests (Method 1313) is indicated by a large circle. Method 1313 data is used to determine the available content (i.e., the fraction of total content that is available for leaching under environmental conditions), as the mass release in mg/kg-dry associated with the maximum COPC concentration at pH 2, 9, or 13 (Rivera et al., 2016; U.S. EPA, 2019a). Percolation

column test (Method 1314) data indicates the evolution of equilibrium concentrations with increasing cumulative L/S ($\Sigma L/S$), e.g., with continued infiltration. The Method 1315 data shows the cumulative mass release of COPCs normalized to the surface area of test material exposed to leaching solution. All graphs in Fig. 2 include the method response, method detection limit (MDL), lower limit of quantitation (LLOQs). The assessment threshold also is indicated for Method 1313 and 1314 results. Detailed description of methods and results are provided by Garraabrants et al. (2020).

2.3. Infiltration data for wet and dry environments⁴

Relevant summary data for the precipitation and infiltration (Table 1) was derived (see SI-2) from weather station data (U.S. EPA, 2018) for a relatively wet environment (Nashville, TN) and a relatively dry environment (Pendleton, OR). The annual net infiltration was converted to L/S (L/kg-dry) for field scenarios by multiplying the annual net infiltration, in m/y, by scenario footprint area (400 m²) and dividing the dry mass of the solid in the scenario fill. The annual infiltration for wet (Nashville) and dry (Pendleton) environments represents 0.10 and 0.02 L/kg per year, respectively.

In the percolation assessment, the annual infiltration (L/S per year) was accumulated to generate a cumulative L/S at the end of each time step i , ($\Sigma L/S$) _{i} , which was used in conjunction with percolation column test data to estimate the concentration in COPCs in leachate. In the diffusion scenario, daily infiltration data were separated into “single-day events” (≤ 1 -day) and “extended events” (> 1 -day) with an average extended event duration of 2.3 days for Nashville and 2.5 days for Pendleton. The annual average number of single-day events (N_1) and extended events (N_2), as well as the average infiltration per event (P_1 , P_2), are determined from daily infiltration results. For this illustration, the data in Table 1 were considered constant for each time step (e.g., each year); however, different data could be developed for each time step in the assessment interval.

2.4. Applicable pH domain

Within the LEAF guidance (U.S. EPA, 2019a), a default applicable pH range of 5.5–9.0 is specified for the majority of scenarios and adjustments are recommended to include the material natural pH and the plausible pH domain of the field scenario. The pH domain may be narrowed, for example, to the natural pH ± 1.0 in cases where leachate pH is not influenced by external sources of acidity or alkalinity and the natural pH changes with time are not expected. Similarly, the pH domain may be extended to include the natural pH of alkaline materials and changes in material pH associated with aging. In all cases, the applicable pH domain should be adjusted to the nearest Method 1313 batch extraction pH so that leaching data may be used directly without interpolation.

The applicable pH domain of CFS ($pH_{nat}=6.9$) was defined as $5.5 \leq pH \leq 8.0$, while an applicable pH domain of $8.0 \leq pH \leq 13$ was determined for S/S-CFS ($pH_{nat}=12.8$). These

⁴For this illustration, the evaluation of infiltrating water is estimated using weather station data (U.S. EPA, 2018); however, other approaches for defining infiltration rates (e.g., site-specific measurements) are applicable. Approaches used for regulatory compliance should be verified with the appropriate authority prior to use.

domains reflect the natural pH of both materials and the long-term changes in pH associated with carbonation of S/S materials (Branch et al., 2016; Garrabrants et al., 2004; Gervais et al., 2004; Lin, 2007).

3. Methodology for scenario-based assessments

The general approach for scenario-based assessments is to calculate an estimated average leaching concentration over an assessment interval (C_I^{av}) based on percolation or diffusion scenarios (see flowcharts provided in SI-3). The estimated leaching concentration may be calculated for a single assessment time step (e.g., 1-year) or averaged over several time steps that comprise an assessment interval, I (e.g., yearly time steps over a 30-year assessment):

$$C_I^{av} = \frac{\sum_{i=1}^I C_i^{av}}{I} \quad (1)$$

The assessment is completed by comparing the estimated leaching concentration (C_I^{av}) to a threshold concentration (C_{thres}) in terms of an assessment ratio (AR) that considers the potential for dilution of leachate into groundwater and the attenuation of COPCs through adsorption to surfaces using a dilution attenuation factor (DAF; U.S. EPA, 1995):

$$AR = \frac{C_I^{av}}{C_{thres} \cdot DAF} \quad (2)$$

An assessment ratio less than one ($AR < 1$) indicates that COPC leaching does not exceed the threshold value, while a ratio greater than one ($AR > 1$) indicates that a COPC may remain a potential concern due to leaching. For regulatory programs where DAFs may not be considered under a release scenario (e.g., when the point of compliance is the point of release), the DAF value is set to one. This concentration-based methodology may be readily adapted to regulatory programs where threshold values are based on mass release.

3.1. Leaching concentration for percolation-based scenarios

After each infiltration event, the water content of the soil bed is assumed to return to field capacity and, thus, the volume of leachate generated is roughly equal to the volume of infiltration, assuming no leachate flux from the surface. The leachate is assumed to be in equilibrium with the CFS soil, such that leaching concentrations may be derived directly from LEAF test results. However, the approach for estimating the equilibrium leaching concentration differs depending on whether the measured test concentration is controlled by (i) COPC solubility (i.e., the equilibrium solution is chemically saturated with respect to the COPC) or adsorption/desorption partitioning, or (ii) available content-limited (Garrabrants et al., 2020; U.S. EPA, 2019a).

3.1.1. Solubility-controlled and adsorption/desorption partitioning—For solubility-controlled and adsorption/desorption partitioning, the measured concentration is a strong function of pH and a weak function of L/S (e.g., arsenic in Fig. 1). The estimated

average leachate concentration for an assessment time step (C_i^{av}), is equal to the maximum concentration in Method 1313 taken over the applicable pH domain:

$$C_i^{av} = C_{\max pH \text{ domain}}^{1313} \quad (3)$$

Since concentrations are relatively independent of L/S, this concentration will persist as the $\Sigma L/S$ increases (i.e., subsequent time steps), as long as the pH remains relatively constant (± 0.5 pH unit) and there is sufficient available content to maintain this concentration (Appelo and Postma, 2013; Kosson et al., 2014; Stumm and Morgan, 1996; U.S. EPA, 2014b).

3.1.2. Available content-limited partitioning—When a COPC readily leaches up to its available content and the resulting solution concentration is less than chemical saturation, the measured concentration is a strong function of L/S and a weak function of pH and the leaching behavior is considered limited by the available content of the COPC (U.S. EPA, 2019a). Since infiltrating water displaces porewater as the soil bed returns to field capacity, the displaced leachate is assumed to be at local equilibrium, but this equilibrium concentration can change as $\Sigma L/S$ increases and more-soluble constituents are removed by leaching. Thus, the average leaching concentration for the assessment interval may be approximated using the Method 1314 concentration corresponding to the cumulative L/S at the end of the assessment time step ($\Sigma L/S$)_i:

$$C_i^{av} = C_{(\Sigma L/S)_i}^{1314} \quad (4)$$

An approach for developing average leaching concentrations for intermediate $\Sigma L/S$ using Method 1314 data is described in SI-4.

3.1.3. Depletion of available content—Since the total mass released cannot exceed the available content of a COPC, the cumulative mass at the end of an assessment interval is compared to the available content provided by Method 1313. The interval mass release (R_i) is calculated from the estimated average leaching concentration and the interval L/S (L/S)_i:

$$R_i = C_i^{av} \cdot (L/S)_i \quad (5)$$

The available content remaining in the solid at the end of assessment interval, $(AC)_i$, is estimated from the previous available content value, $(AC)_{i-1}$, and the interval mass release:

$$(AC)_i = (AC)_{i-1} - R_i \quad (6)$$

If the available content for any assessment interval is negative, the COPC has been depleted and the interval mass release is constrained to the previously available content:

$$\text{if } (AC)_i < 0; \text{ then } (AC)_i = 0 \text{ and } R_i = (AC)_{i-1}$$

The estimated average leaching concentration for depleted COPCs is adjusted by rearranging the interval mass release Eq. (5) to $C_i^{av} = R_i / \left(\frac{L}{s} \right)_i$. For all subsequent intervals, the estimated leaching concentration is zero since no leachable COPCs remains in the solid.

3.2. Leaching concentration for diffusion scenarios

Low permeability materials deposited in the vadose zone are exposed to intermittent convection of contact water with each infiltration event such that internal gradients can relax (i.e., decrease) between events as COPCs at the interface diffuse into a relative stagnant zone. For the equivalent amount of contact water, cumulative mass release under intermittent leaching/stagnant zone conditions is greater than continuous leaching due to increased COPC concentrations at the interface after internal gradient relaxation (Garrabrants et al., 2002; Sanchez et al., 2003). Thus, there is essentially a cyclic behavior to mass release for each infiltration event based on the duration of the infiltration event similar to the beginning leaching intervals of Method 1315. The measured cumulative release in Method 1315 testing through 2 days of leaching was considered to estimate the cumulative release of COPCs during infiltration events lasting more than 2 days because of the rapid decrease in COPC flux with leaching time (Fig. 2). This approximation is supported by the average extended event duration of 2.3 and 2.5 days for the wet (Nashville) and dry (Pendleton) environments (Table 1). Additionally, release estimate from mass transfer rate leaching tests tend to be conservative due to the much greater liquid-to-surface-area compared to field conditions where large fills are exposed to limited infiltration. Table 2.

The methodology for estimating leaching concentrations under intermittent flow is based on the approach used to estimate COPC release from cement pavements containing coal combustion fly ash (U.S. EPA, 2014a). An effective leaching concentration is calculated for both 1-day and 2-day infiltration events as the estimated COPC mass released from the monolith dissolved in the volume of contacting water. The estimated average leaching concentration over the assessment interval is determined as the weighted average of single-day and extended infiltration events.

3.2.1. Effective concentrations for 1-day and 2-day events—The effective concentration for single-day infiltration events (<1-day) is estimated using cumulative release data from the mass transfer rate test (SI-5). The cumulative mass release at the end of a Method 1315 leaching interval, J , is calculated from the interval concentration data (C_j^{1315}), the volume of eluate collected in each interval (V_j^{1315}) and the exposed surface area of the test sample (A_{exp}^{1315}):

$$\Sigma R_J = \frac{\sum_{j=1}^J (C_j^{1315} \cdot V_j^{1315})}{A_{exp}^{1315}} \quad (7)$$

The mass released for the 1-day interval is represented by the difference in cumulative release between the first leaching interval (ΣR_1 , at cumulative leaching time of 0.08 days) and second leaching interval (ΣR_2 , at cumulative leaching of 1.1 days).⁵ The 1-day mass release for the scenario ($M1$) is obtained from Method 1315 data as $\Sigma R_2 - \Sigma R_1$ and is scaled

to the exposed surface area of the disposed material (A_{exp}). The volume of contact water for a 1-day event ($V1$) is the average infiltration per 1-day event ($P1$) multiplied by the infiltration-impacted area (A_{inf}). The effective concentration of a COPC for a 1-day event may be estimated as:

$$C1 = \frac{M1}{V1} = \frac{(\Sigma R_2 - \Sigma R_1) \cdot A_{exp}}{P1 \cdot A_{inf}} \left(\frac{m^3}{1000 L} \right) \quad (8)$$

Mass release from Method 1315 over a 2-day cumulative leaching interval was considered a good approximation for release over extended infiltration events > 1 day. An analogous equation for a 2-day infiltration event uses a Method 1315 mass release ($\Sigma R_3 - \Sigma R_1$) and the average volume of infiltration over a 2-day event ($P2$):

$$C2 = \frac{M2}{V2} = \frac{(\Sigma R_3 - \Sigma R_1) \cdot A_{exp}}{P2 \cdot A_{inf}} \left(\frac{m^3}{1000 L} \right) \quad (9)$$

3.2.2. Equilibrium limitations—The effective leaching concentrations for 1-day and 2-day infiltration events are based on Method 1315 data generated from a relatively small sample exposed to a large bath of water to maximize the internal concentration gradients that drive mass transport. However, these conditions are not representative of the majority of subsurface field conditions where, typically, a small volume of water contacts a relatively large surface area.⁶ Thus, the effective concentration calculations for 1-day and 2-day infiltration events can numerically exceed the thermodynamic bounds of chemical equilibrium (i.e., as represented by the Method 1313 data). Therefore, if the effective leaching concentration for any 1-day or 2-day event is greater than the measured Method 1313 solubility-controlled eluate concentration, the effective concentration should be set to the Method 1313 maximum concentration over the pH domain:

$$\text{if } C1(\text{or } C2) > C_{maxpH domain}^{1313}; \text{ then } C1(\text{or } C2) = C_{maxpH domain}^{1313}$$

3.2.3. Estimated average leaching concentration—An average concentration for an assessment interval, i , may be calculated from the number of single-day and extended infiltration events ($N1, N2$) and the estimated leaching concentrations ($C1, C2$) as a weighted average concentration over the series of 1- and 2-day infiltration events:

$$C_i^{av} = \left[\frac{N1 \cdot C1 + N2 \cdot C2}{N1 + N2} \right] \quad (10)$$

⁵The concentration measured in the first interval of Method 1315 is often biased by surface dissolution and sample preparation (e.g., wash-off of cutting swarf). Therefore, the mass released during this first interval with eluate exchange at 0.08 days is not used in calculations since it is not representative of the leaching behavior of the bulk material.

⁶The rate mass transport by diffusion and dispersion orthogonal to the monolith surface is relatively slow compared to mass transport in the flow direction (assuming no-flow boundary condition at the monolith surface), resulting in a water film effect under normal infiltration velocities, unless there is preferential flow along the monolith-soil or monolith-backfill interface.

3.2.4. Depletion of available content—The diffusion scenario has a similar restriction as in the percolation scenario that the total mass released cannot exceed the available content for a COPC. The interval mass release for an assessment interval $i(R_i)$ is calculated from the total mass from 1-day events ($\Sigma M1$) and 2-day events ($\Sigma M2$) normalized to the dry mass of material in the scenario (m_d) as:

$$R_i = \frac{M1}{m_d} + \frac{M2}{m_d} = \left[\frac{(N1 \cdot C1 \cdot P1 \cdot A_{inf})}{m_d} + \frac{(N2 \cdot C2 \cdot P2 \cdot A_{inf})}{m_d} \right] \left(\frac{1000L}{m^3} \right) \quad (11)$$

The available content at the end of an assessment interval can be estimated as the previous interval available content minus the mass released over the current assessment interval using Eq. 6. If the available content for any assessment interval is less than zero, the COPC has depleted and the interval mass release is limited to the available content at the end of the previous interval:

$$\text{if } (AC)_i < 0; \text{ then } (AC)_i = 0 \text{ and } M_i = (AC)_{i-1}$$

The average leaching concentration for each COPC should be adjusted to match the mass released over the assessment interval. Analogous to the percolation scenario, ARs may be calculated for the mass transport scenario based on the estimated leaching concentration for an assessment interval or an averaged value for an assessment period.

4. Leaching assessment results

The final step for each scenario assessment is to generate ARs based on threshold criteria and DAF values. The estimated leaching concentration and ARs for 1-year, 5-year, and 30-year assessment periods with DAF = 1 are calculated for CFS in a percolation scenario (Table 3) and for S/S-CFS in diffusion scenario (Table 4). ARs shown in bold italics ($AR \leq 1$) indicate COPCs that are not expected to leach at concentrations greater than the threshold. The S/S treatment of smelter soil generally decreases COPC leaching significantly for all assessment periods, resulting in a leaching reduction of about an order-of-magnitude for most COPCs. The reduction with treatment for cadmium is dramatic with ARs of > 1000 for CFS and 0.3 for S/S-CFS. In comparison, the reduction for arsenic is about a factor of 2 because of similar equilibrium concentrations near natural pH of the two materials (Fig. 2).

4.1. Effect of assessment period

When the average AR for 1-year and 30-year assessment period are compared, the results for solubility-controlled COPCs (As, Cd, Cr, Cu, F^- , Pb, Sb, Tl, Zn) remain constant. These COPCs are expected to leach at pH-dependent equilibrium until the available content is depleted, which does not occur over the 30-year assessment period. For available content-limited COPCs (Cl^- , NO_3^- , Se, SO_4^{2-}), the ARs for the 1-year assessment are greater than those for the 30-year assessment because the estimated leaching concentration is a strong function of cumulative L/S with peak leaching concentrations during early assessment

intervals. The ARs for NO_3^- are reduced significantly in the 30-year assessment period due to rapid depletion of the available content after two years.

4.2. Comparison of wet and dry environments

The scenario assessment results compared for wet and dry environments over a 30-year assessment interval (Table 5) show that ARs for available content-limited COPCs (Cl^- , NO_3^- , Se, SO_4^{2-}) in percolation scenarios were 3–7 times greater in dry environments than wet environments. The increase in concentration for dry environments is primarily due to the decrease in leaching volume ($\Sigma L/S$) over a given year. For solubility-controlled COPCs, the percolation assessment results remained unchanged because leaching concentrations are based on pH-dependence rather than L/S-dependence. Thus, although the mass of COPC released may be less in a dry environment relative to a wet environment, leaching concentrations may increase for some COPCs.

In diffusion scenarios, the dry environment showed 50% less COPC mass leaching than the wet environment for 1-day and 2-day infiltration events and the total amount of infiltration per event (Table 1). The estimated average leaching concentrations of most COPCs increased for a given assessment interval under drier conditions. Chloride and nitrate leaching concentrations (C_i^{av}) did not increase because the calculated effective concentrations from diffusion (C_1 , C_2) were greater than the equilibrium concentration over the applicable pH domain ($C_{maxpH\text{domain}}^{1313}$), and therefore the effective concentrations were reset to the corresponding equilibrium values. For the remaining COPCs, the ARs in the dry environment are 2.7 times greater than in the wet environment resulting in some COPCs with ARs ≤ 1 for the wet location (Cd, Sb, Se, SO_4^{2-}), but ARs > 1 for the dry location.

The mass release of selected COPCs from CFS in a percolation scenario (red) and S/S-CFS in a diffusion scenario (blue) is presented in Fig. 3. Comparisons for all COPCs in the scenario assessment and process for normalizing release for percolation scenarios (mg/kg-dry) and diffusion scenarios (mg/m²) for comparison are presented in SI-6. The solid lines indicate results from the wet environment while dashed lines indicate the dry environment. For all COPCs, the calculated cumulative release is lower when the material is subjected to the dry environment, regardless of the material or scenario. However, the reduction generally is less than an order-of-magnitude for most COPCs.

4.3. Assessment of treatment effectiveness

When leaching from percolation and diffusion scenarios are compared, ARs provide a uniform basis for comparison and for evaluation of treatment effectiveness. Scenario-based assessments also provide time-dependent evaluations that include rates of release based on the annual rate of infiltration and depletion of COPCs.

The cumulative release of selected COPCs are provided for untreated CFS and cement-treated S/S-CFS in the wet environment (Fig. 4 and SI-7). In each figure, the available content is shown as the upper limit of mass release and the CFS mass release was normalized to a mass value using scenario data (see SI-6). These time-based evaluations are particularly useful for graphically demonstrating the effectiveness of a treatment process,

especially when more than one scenario is required (e.g., when treatment changes the physical nature of the subject material). These comparisons show that treatment of S/S-CFS material would be expected to reduce the rate of leaching of Cd, F⁻, Sb, Se, SO₄²⁻, Tl and Zn by an order-of-magnitude or more over the 30-year assessment period. Although a reduction in cumulative release was observed for arsenic and copper, the effect was not as strong as for the other COPCs. The shape of the cumulative release curves for available content-limited COPCs, chloride and nitrate, in particular, indicate how the dependence of the percolation scenario on the ΣL/S results in a flattening of the leaching curves.

5. Conclusions

This scenario assessment methodology provides a consistent, robust approach for estimating potential leaching concentrations by incorporating LEAF leaching tests results with conceptual release models and scenario-specific conditions. The methodology distinguishes percolation scenarios for granular materials (e.g., soils, sediments) and diffusion scenarios for low permeability or monolithic materials (e.g., cements, clays).

All leaching assessments make simplifying assumptions that add uncertainty. In this study, net infiltration parameters were approximated from weather station data and considered constant for each assessment interval; however, infiltration data derived by other methods (e.g., site-specific recharge rates that vary yearly) may further refine the assessment. The presented methodology addresses long-term effects of aging (e.g., carbonation of cement-based materials) through adjustment of the applicable pH domains of the assessment scenario. Thus, the presented methodology provides upper-bound estimates of COPC leaching that are sensitive to the physical and chemical characteristics of the subject material and the scenario conditions.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments and disclaimer

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Nomenclature

Variable

A_{exp}^{1315}	Method 1315 exposed surface area of test sample (m ² _{sample} ; Eq. (7))
A_{exp}	exposed surface area of the material in scenario (m ² _{mat} ; Eqs. (8 and 9))

A_{inf}	plan view area of infiltrating water (m^2_{water} ; Eqs. (8 and 9))
$(AC)_i$	available content at end of assessment interval i (mg/kg; Eq. (6))
$(AC)_{i-1}$	available content at end of assessment interval $i - 1$ (mg/kg; Eq. (6))
AR	assessment ratio for assessment period I (2)
C_i^{av}	estimated average leaching concentration for assessment interval, i (mg/L; Eqs. (1, 3, 4, 5, 10))
C_I^{av}	estimated average leaching concentration over assessment interval I (mg/L; Eqs. (1 and 2))
$C_{maxpH domain}^{1313}$	Method 1313 maximum concentration over applicable pH domain (mg/L; Eq. (3))
$C_{(\Sigma L/S)_i}^{1314}$	Method 1314 concentration at $(\Sigma L/S)_i$ (mg/L; Eq. (4))
C_j^{1315}	Method 1315 concentration for test interval j (mg/L; Eq. (7))
C_{thres}	threshold concentration (mg/L; Eq. (2))
$C1$	effective concentration – 1-day infiltration (mg/L per event; Eqs. (8, 10 and 11))
$C2$	effective concentration – 2-day infiltration (mg/L per event; Eqs. (9–11))
DAF	dilution and attenuation factor (2)
$(L/S)_i$	liquid-to-solid ratio for assessment interval i (L/kg-dry; Eq. (5))
m_d	dry mass of material (soil bed or S/S) (kg-dry; Eq. (11))
$M1$	mass release for a 1-day infiltration event (mg; Eq. (8))
$M2$	mass release for a 2-day infiltration event (mg; Eq. (9))
$N1$	number of infiltration events ≤ 1 day (events; Eq. (10))
$N2$	number of infiltration events > 1 day (events; Eq. (10))
$P1$	net infiltration for events ≤ 1 day (m; Eq. (8))
$P1$	net infiltration for events > 1 day (m; Eq. (9))
R_i	mass released in assessment interval i (mg/kg-dry; Eqs. (5, 6 and 11))

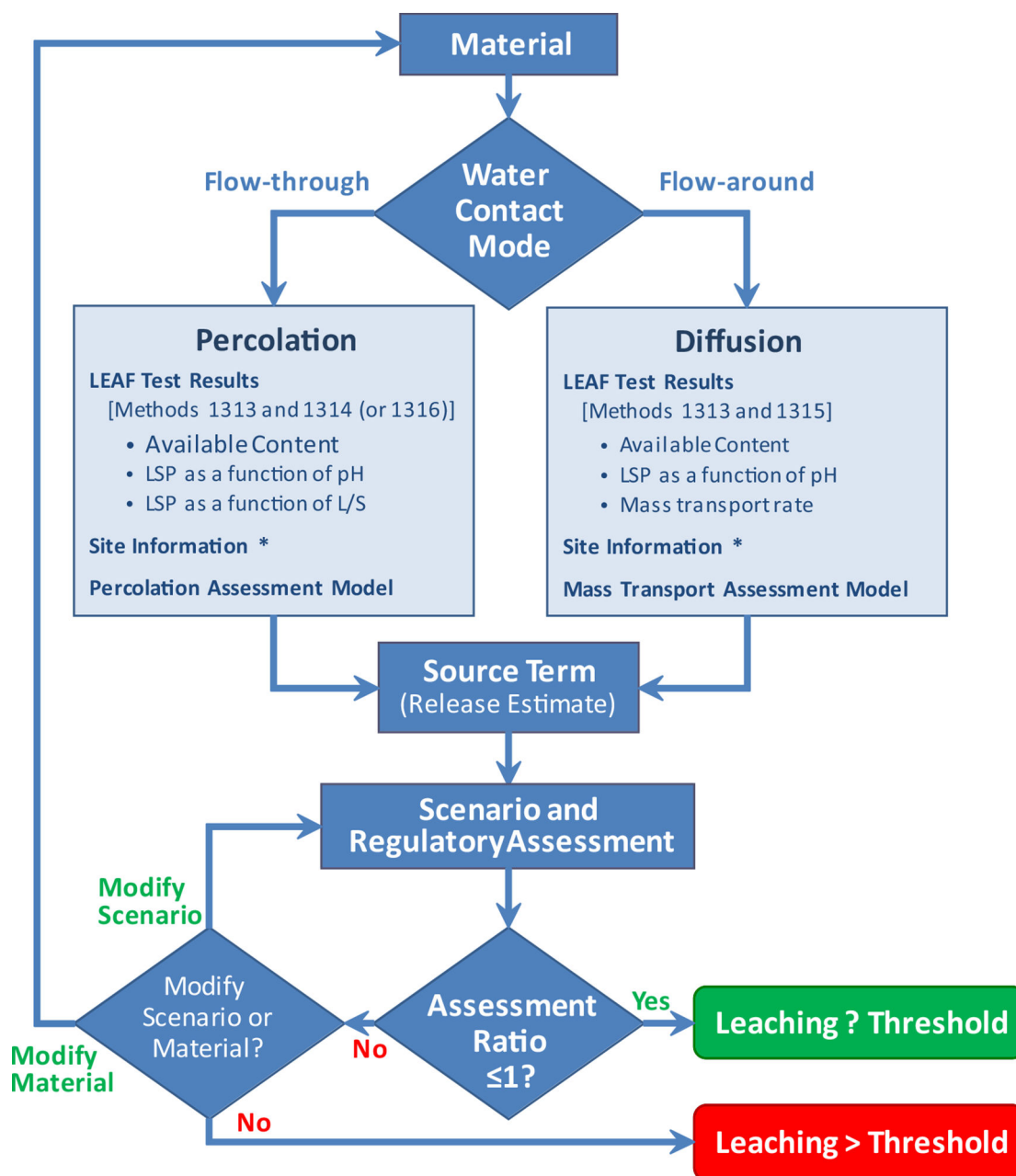
V1	contact water volume for a 1-day infiltration event (L; Eq. (8))
V2	contact water volume for a 2-day infiltration event (L; Eq. (9))
V_j^{1315}	Method 1315 eluate volume for test interval j (L; Eq. (7))
$\Sigma M1$	total mass release for all 1-day events in an interval (mg; Eq. (11))
$\Sigma M2$	total mass release for all 2-day events in an interval (mg; Eq. (11))
ΣR_j	Method 1315 cumulative release for interval J (mg/m ² _{mat} ; Eq. (7))
ΣR_1	Method 1315 cumulative release – 1st interval (mg/m ² _{mat} ; Eqs. (8 and 9))
ΣR_2	Method 1315 cumulative release – 2nd interval (mg/m ² _{mat} ; Eq. (8))
ΣR_3	Method 1315 cumulative release – 3rd interval (mg/m ² _{mat} ; Eq. (9))

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* Default values or site-specific information
Figure modified from U.S. EPA (2019a).

Fig. 1.
Scenario-based assessment approach based on LEAF guidance (U.S. EPA, 2019a).

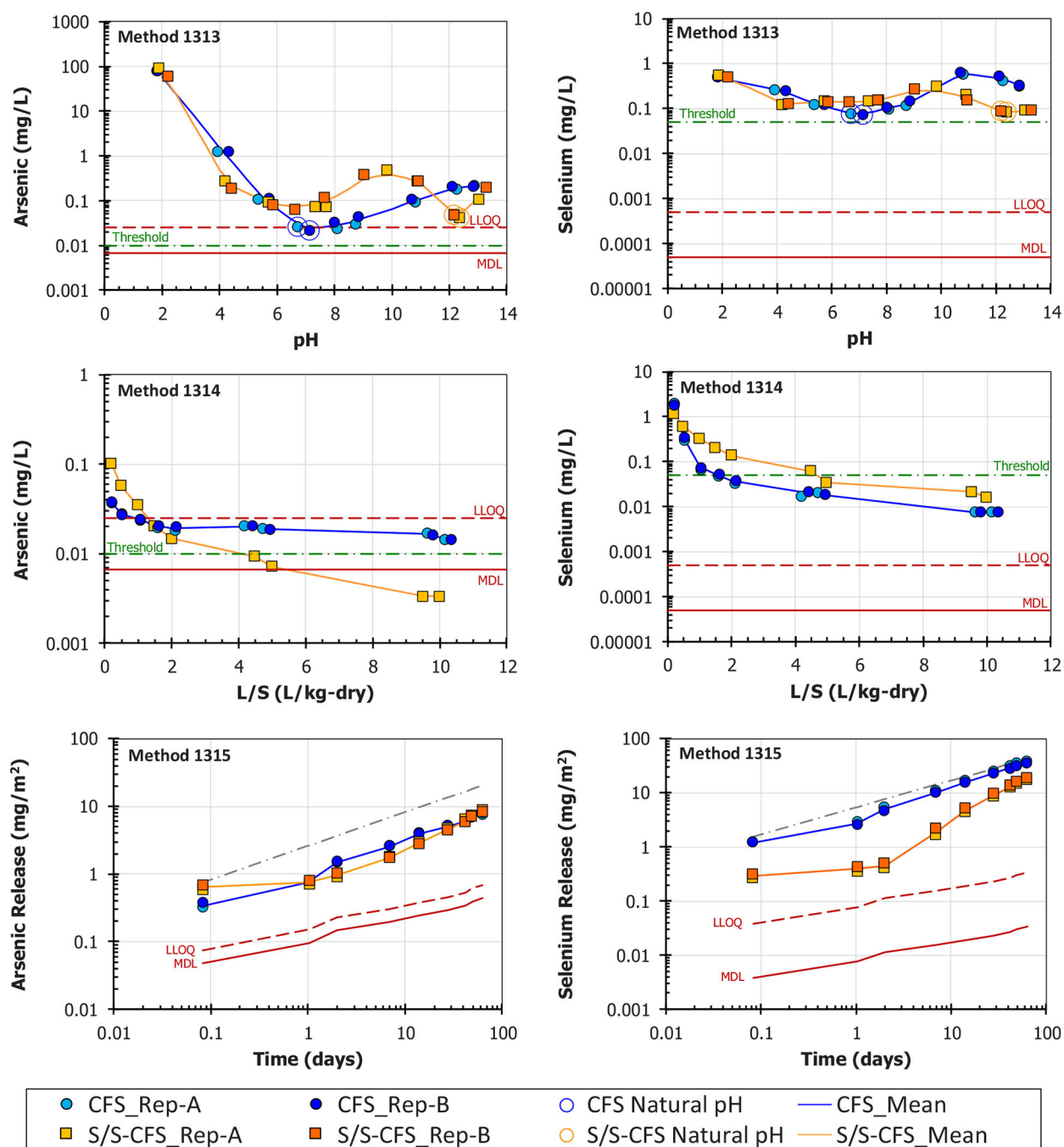


Fig. 2.

Example of LEAF test data used to support scenario-based leaching assessments: Method 1313 (top), Method 1314 (middle), and Method 1315 (bottom).uplicate data shown for CFS (gold/orange) and S/S-CFS (light blue/blue).

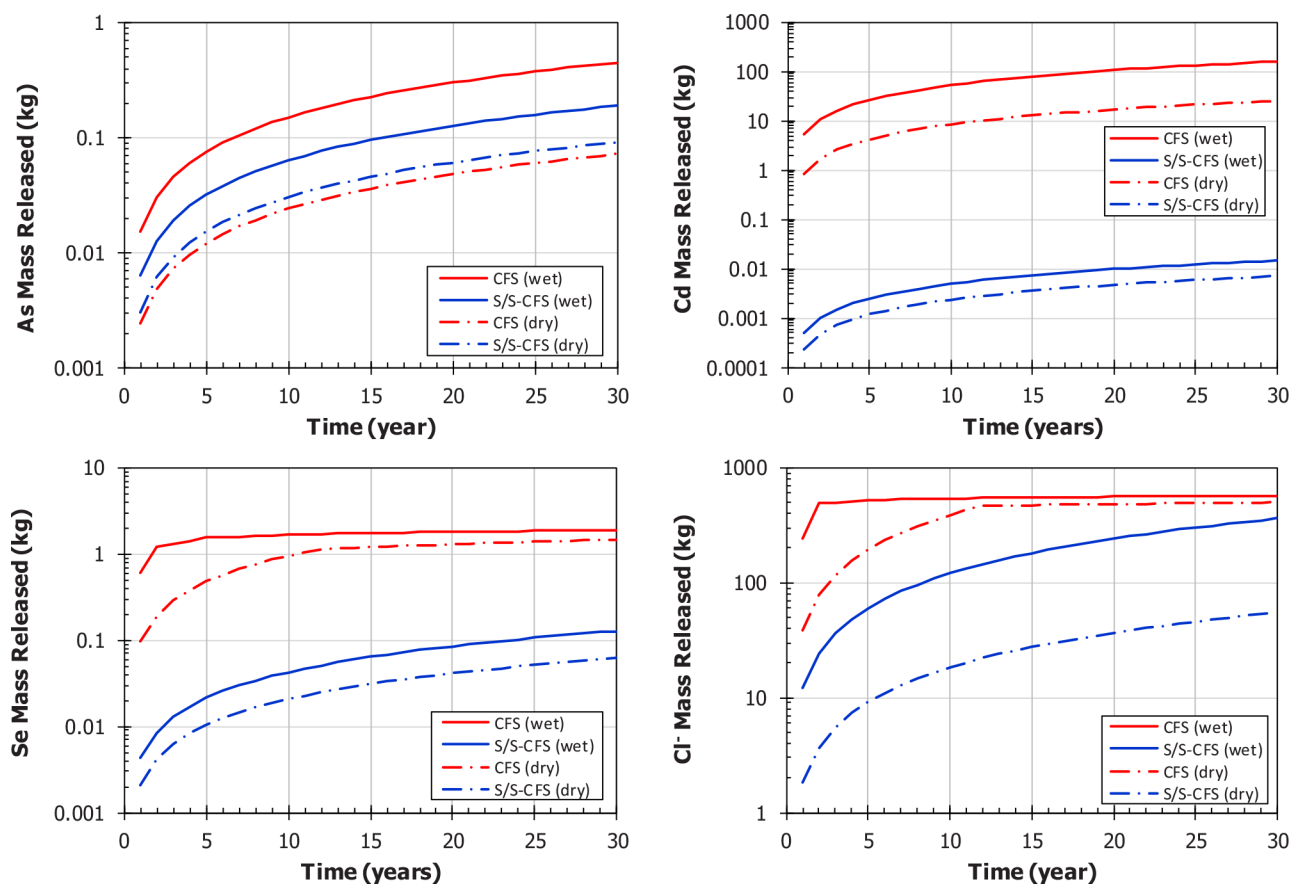


Fig. 3.
The effect of wet (Nashville, TN) and dry (Pendleton, OR) environments on cumulative release of a 30-year assessment period.

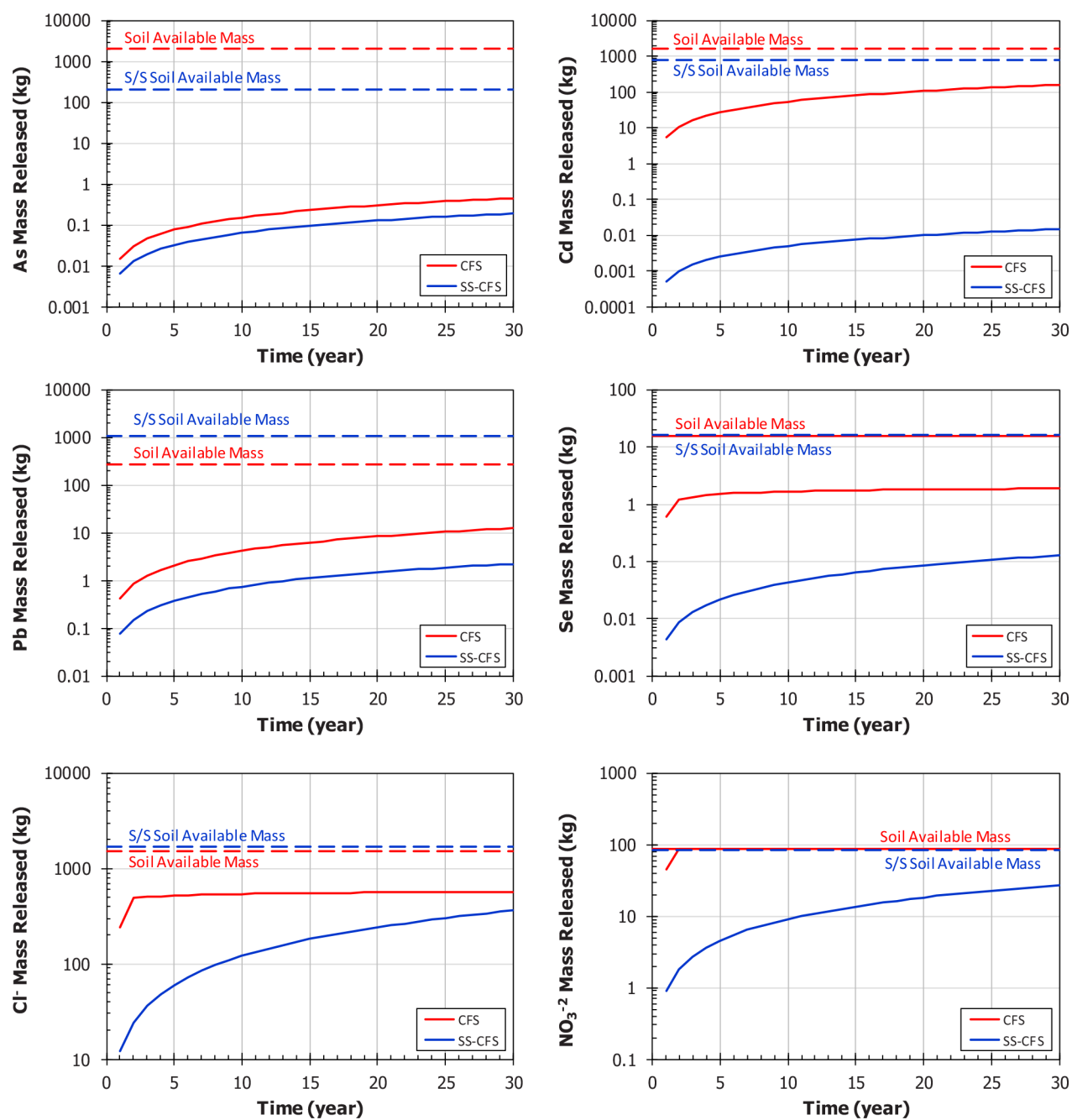


Fig. 4.
Treatment effectiveness through comparison of cumulative release over a 30-year assessment period.

Table 1

Average precipitation and infiltration for wet (Nashville, TN) and dry (Pendleton, OR) environments (1961–1990).

	Units	Wet	Dry
City, State		Nashville, TN	Pendleton, OR
Annual Precipitation	cm/y	120	31
Days with Precipitation	d/y	118	96
Annual Net Infiltration	cm/y	82	13
Infiltration Events ≤ 1 -day (N1)	events/y	32	16
Infiltration Events > 1 -day (N2)	events/y	13	6
Net Infiltration of Events ≤ 1 -day (P1)	cm	1.2	0.38
Net Infiltration of Events > 1 -day (P2)	cm	3.5	1.1
Infiltration Events > 2 -days	%	6.7%	9.5%

Table 2

Scenario-based assessment results for 1-, 5-, and 30-year assessments of CFS in a percolation scenario for a wet environment (Nashville, TN). Bold, italicized text indicates ARs ≤ 1 (i.e., no further assessment is required).

COPC	Threshold (mg/L)	1-Year Assessment		5-Year Assessment		30-Year Assessment	
		C _{av} (mg/L)	AR (-)	C _{av} (mg/L)	AR (-)	C _{av} (mg/L)	AR (-)
Antimony	0.006	0.091	15	0.091	15	0.091	15
Arsenic	0.01	0.046	4.6	0.046	4.6	0.046	4.6
Cadmium	0.005	16	>1000	16	>1000	16	>1000
Chloride	250 *	740	3.0	320	1.3	58	0.2
Chromium	0.1	–	–	–	–	–	–
Copper	1.3	2.8	2.2	2.8	2.2	2.8	2.2
Fluoride	4	2.6	0.6	2.6	0.6	2.6	0.6
Lead	0.015	1.3	85	1.3	85	1.3	85
Nitrate	44	137	3.1	53	1.2	8.8	0.2
Selenium	0.05	1.8	36	0.94	19	0.19	3.8
Sulfate	250 *	12,000	49	7200	29	2900	12
Thallium	0.002	1.9	940	1.9	940	1.9	940
Zinc	5 *	67	14	67	14	67	14

Notes: Threshold values based on U.S. EPA National Primary Drinking Water Regulations (U.S. EPA, 2009) with * indicating secondary drinking water criteria.

(-) No percolation scenario analysis was conducted for chromium since screening-level ARs were less than 1 (Garraabrants et al., 2020).

Table 3

Scenario-based assessment results for 1-, 5-, and 30-year assessments of S/S-CFS in a diffusion scenario for a wet environment (Nashville, TN). Bold, italicized text indicates ARs ≤ 1 (i.e., no further assessment is required).

COPC	Threshold (mg/L)	1-Year Assessment		5-Year Assessment		30-Year Assessment	
		C _{av} (mg/L)	AR (-)	C _{av} (mg/L)	AR (-)	C _{av} (mg/L)	AR (-)
Antimony	0.006	0.0022	0.4	0.0022	0.4	0.0022	0.4
Arsenic	0.01	0.019	1.9	0.019	1.9	0.019	1.9
Cadmium	0.005	0.0015	0.3	0.0015	0.3	0.0015	0.3
Chloride	250 *	36	0.1	36	0.1	36	0.1
Chromium	0.1	0.015	0.2	0.015	0.2	0.015	0.2
Copper	1.3	0.072	0.06	0.072	0.06	0.072	0.06
Fluoride	4	0.020	<0.01	0.020	<0.01	0.019	<0.01
Lead	0.015	0.21	14	0.21	14	0.21	14
Nitrate	44	2.7	0.06	2.7	0.06	2.7	0.06
Selenium	0.05	0.014	0.3	0.014	0.3	0.014	0.3
Sulfate	250 *	150	0.6	150	0.6	150	0.6
Thallium	0.002	0.18	91	0.18	91	0.18	91
Zinc	5 *	0.019	<0.01	0.019	<0.01	0.019	<0.01

Notes: Threshold values based on U.S. EPA National Primary Drinking Water Regulations (U.S. EPA, 2009) with * indicating secondary drinking water criteria.

Table 4

Comparison 30-year assessments in wet (Nashville, TN) and dry (Pendleton, OR) environments for CFS (percolation scenario) and S/S-CFS (diffusion scenario). Bold italicized text indicates ARs ≤ 1 (i.e., no further assessment is required).

COPC	Threshold (mg/L)	CFS in Percolation Scenario			S/S-CFS in Mass Transport Scenario					
		Nashville, TN (wet)		Pendleton, OR (dry)		Nashville, TN (wet)		Pendleton, OR (dry)		
		C _{av} (mg/L)	AR (-)	C _{av} (mg/L)	AR (-)	C _{av} (mg/L)	AR (-)	C _{av} (mg/L)	AR (-)	AR (-)
Antimony	0.006	0.091	15	0.091	15	0.0022	0.4	0.0072	1.2	
Arsenic	0.01	0.046	4.6	0.046	4.6	0.019	1.9	0.060	6.0	
Cadmium	0.005	16	>1000	16	>1000	0.0015	0.3	0.0047	0.9	
Chloride	250 *	58	0.2	320	1.3	36	0.1	36	0.1	
Chromium	0.1	–	–	–	–	0.015	0.2	0.048	0.5	
Copper	1.3	2.8	2.2	2.8	2.2	0.072	0.06	0.23	0.2	
Fluoride	4	2.6	0.6	2.6	0.6	0.019	< 0.01	0.064	0.02	
Lead	0.015	1.3	85	1.3	85	0.21	14	0.67	45	
Nitrate	44	8.8	0.2	55	1.2	2.7	0.06	2.7	0.06	
Selenium	0.05	0.19	3.8	0.9	19	0.014	0.3	0.046	0.9	
Sulfate	250 *	2900	12	7200	29	150	0.6	470	1.9	
Thallium	0.002	1.9	940	1.9	940	0.18	91	0.23	120	
Zinc	5 *	67	14	67	14	0.019	< 0.01	0.061	0.01	

Notes: Threshold values based on U.S. EPA National Primary Drinking Water Regulations (U.S. EPA, 2009) with * indicating secondary drinking water criteria.

(-) No percolation scenario analysis was conducted for chromium since screening-level ARs were less than 1 (Garraabrants et al., 2020).